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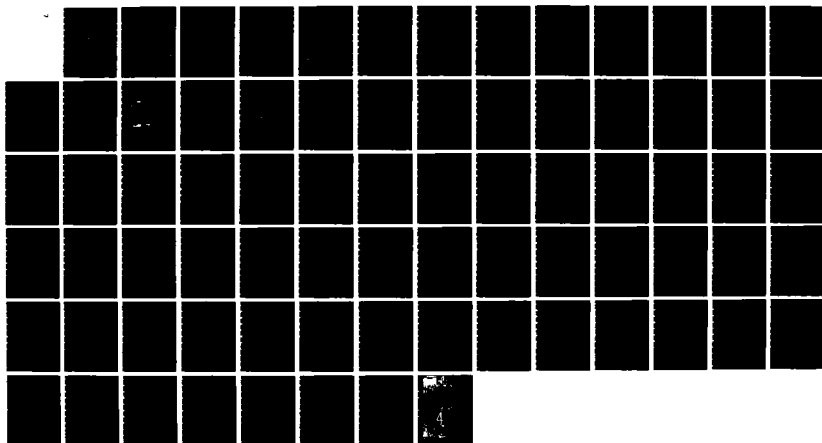
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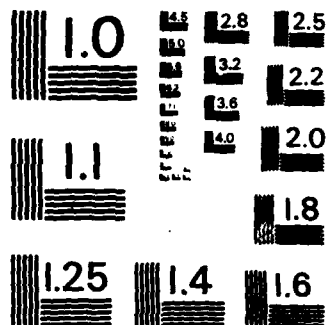
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U.S. Department  
of Transportation  
Federal Aviation  
Administration

# Cruise Aircraft Effects, 1981 Status

Office of Environment  
and Energy  
Washington, D.C. 20591

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Robert C. Oliver

January 1982

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# PREFACE

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# ABSTRACT

Recent (post-1978) developments are discussed in the longstanding question of possible environmental effects of aircraft emissions at cruise altitudes. Computer models of atmospheric ozone, using recently accepted changes in certain key reaction rates now again indicate that  $\text{NO}_x$  from SSTs at 17-20 km will reduce the ozone column, as was the case about 1974. The same chemistry changes have materially lessened the computed effects of halocarbons. No recent results on effects of  $\text{NO}_x$  from subsonic aircraft (which fly at altitudes of 6-14 km) are available, but minor ozone column enhancement is still expected. Changes are being considered in mathematical representations of the transport process in one-dimensional (1-D) parameterizations, which may reduce the spread of effects computed by different groups. A coherent picture may be developing on aircraft effects which will require further study to establish and to quantify. Other issues (effects of climate, contrails) are also discussed.

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## SUMMARY

Available information, emphasizing recent (post-1978) developments, is here summarized with regard to the effects aircraft emissions (primarily the  $\text{NO}_x$  and  $\text{H}_2\text{O}$  constituents) may have on the earth's ozone shield; some discussion of climatic effects is also included. Sources are cited in the text. The following topics are of interest:

Fleet Projections (Section 2.2). Air traffic projections for 1990 are now such that the "high" case developed in 1976 seems definitely unrealistic. The 1976 "base" case is suggested as a preferable case for modeling exercises, and an emission inventory included herein. New projections are desirable. No projection for a 1990 SST fleet is very credible, because continued operation of the few SSTs now in service appears problematical.

$\text{NO}_x$  Emissions (Section 2.3). The longstanding question of the discrepancy between spectroscopically-determined (via ultraviolet-spectroscopy) and probe-determined  $\text{NO}_x$  emission-indices has been partly resolved, through corrections in the ultraviolet (UV) technique. It is now agreed that the two techniques give essentially the same results in non-reacting subsonic streams. There is still disagreement for reacting (particularly supersonic) streams. The difficulties with the UV technique, and the need for accurate velocity and temperature data inside the stream make the UV technique less attractive than was initially apparent. The issues involved still need further resolution.

Ozone Effects (Section 3). The computed effects on the ozone column of the  $\text{NO}_x$  emissions from large fleets of SSTs have continued to change, and are again approaching the depletion effects computed in the Climatic Impact Assessment Program in 1974. This major change, down from an enhancement in the column computed in 1978 and 1979, is due primarily to changes in the accepted reaction rates of OH with  $\text{HNO}_3$

and  $\text{HO}_2\text{NO}_2$ . The Lawrence Livermore National Laboratory (LLNL) one-dimensional (1-D) model with the Chang 1976  $\text{K}_z$  profile shows 7.1 percent depletion of ozone with 2000 molecules  $\text{-cm}^{-3}\text{-sec}^{-1}$  over 1 km at 20 km, and 2.2 percent depletion with the same injection at 17 km.

$\text{K}_z$  revisions, which may be called for, lead to larger calculated depletions. If taken as a global average, this injection rate corresponds to  $2.46 \times 10^9$  kg  $\text{NO}_2/\text{yr}$ , or at typical emission indices, about  $1.4 \times 10^{11}$  kg fuel/yr, which is of the order of total current jet fuel consumption. The corresponding computed 1-D ozone depletion values in CIAP, with the LLNL 1-D model in use at that time were about 10 and 5 percent. Other models gave and would still be expected to give, larger values. A recent 2-D model study at NASA-Ames is also cited, which shows about 1 percent depletion for a fleet of 250 SSTs ( $1.5 \times 10^8$  kg/yr as  $\text{NO}_2$ ) flying at an altitude of 20 km, using advanced low-emission engines.

Effects of SST water vapor emissions are debatable but small. The 2-D model study cited above showed that water emissions of the fleet would reduce the ozone column by about 0.5 percent. However, 1-D model studies at LLNL have shown the effects of water vapor to be reduced (in absolute value) if thermal reequilibration and hydrostatic readjustment are included.

It should be noted that computed SST  $\text{NO}_x$  effects on the ozone column depend, for any given set of reactions and reaction rates and the model parameters, on the composition of the reference atmosphere. The composition will change with time; hence, SST  $\text{NO}_x$  effects on ozone will depend on when such a fleet flies, a point noted in the 2-D model studies cited above. Increased chlorine content reduces  $\text{NO}_x$  effects, and vice versa, but not equally. The non-additivity of effects makes particularly difficult the statement of effects of other perturbing influences (such as effects of chlorofluorocarbons) which will not reach equilibrium, even at fixed emission rates, for many decades, over which period there will be many other changes, as in  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ , etc. To illustrate, using recent LLNL 1-D model results, the ozone depletion at steady-state from current halocarbon releases is 5 percent (significantly lower than earlier values, which reached 19 percent in 1979) if the current atmospheric composition is otherwise maintained. Similarly, if  $\text{N}_2\text{O}$  is doubled, the computed decrease in ozone is 12.4 percent. Should both occur together, the total depletion is 12.9 percent, showing

that the two effects are far from additive.  $\text{CO}_2$  increases have a positive influence on the ozone column, reducing such depletions. Reference atmospheres should be stated when citing computed effects.

Up-to-date calculations of subsonic fleet effects are not available, but a minor increase in the ozone column is still expected. The combined effects on a global average (1-D) basis over the 1970 to 1980 period of  $\text{CO}_2$  increases, halocarbon increases, and subsonic fleet  $\text{NO}_x$  increases is apparently near zero. Because subsonic effects are more localized than are halocarbon effects, uniform compensation cannot be expected. Uncertainties are still substantial.

Model Validity (Sections 4.1, 4.2). Revisions in 1-D  $K_z$  profiles have been suggested which may reduce discrepancies between different groups. Recent changes in reaction kinetics have minimized certain discrepancies in measurements, which implied that model OH concentrations in the lower stratosphere were excessively high. These same changes also lead to increased calculated effects of SST  $\text{NO}_x$  injections, and presumably would bring computed results of the nuclear weapons tests in the early 1960's to or near the 4 to 8 percent depletion in the Northern Hemisphere calculated about 1975. The calculations have not been rerun. Observed ozone effects are claimed to be consistent with somewhat smaller (2 to 4.5 percent) maximum effects; the differences are within the uncertainty in source strength. The solar proton event of August 1972 has been re-examined, with reasonable agreement now apparent between theory and measurement in a regime where transport is not very important. A computation of the effects resulting from meteorite entry, which may explain the 1908 Siberian Tunguska event, suggest that depletions of 30 percent  $\pm$  15 percent resulted, due to a fivefold temporary increase in total stratospheric odd nitrogen. The Smithsonian observations of the time, which rather crudely permit ozone column estimation (using Chappuis absorption) seem to be consistent with the computations. An interpretation of satellite data suggests that observed ozone decreases in the 30 to 50 km region of the order of 0.5 percent per year in the 1970 to 1980 period are consistent with calculated effects due to halocarbons.

Unfortunately, none of these observations permit confident assertion that current models for steady-state injections of  $\text{NO}_x$  at 17 to 20 km are "correct" or valid. This is due not only to continuing uncertainties in reaction rates, but also to questions on model representation of transport processes: transport processes are time- and scale-dependent, making comparison of pulsed and steady-state perturbations difficult. Seasonal factors would enter in the region where SST traffic would be concentrated (17 to 20 km at latitudes of 40 to 60°N). Neither scale-dependent nor seasonal factors can be included in 1-D models.

Despite these continuing difficulties, much progress has been made. It may even be that a coherent picture is emerging which will stand up over time. Continued effort is required if confident action (or inaction) based on forecast effects is to be carried out.

Other Effects (Sections 4.3 and 4.4). Little is new in the skin cancer area. A discussion is included of a recent study indicating that contrails from subsonic aircraft in heavily traveled regions in the Midwest may cause increased cloudiness, reducing differences between night and day temperatures and increasing rainfall. The effects are not large, and are confounded by other trends, such as industrial emissions. The cited effects may be advantageous to agriculture. Such effects are almost certainly negligible on a global scale at current and foreseeable traffic levels.

## 1.0 INTRODUCTION

This paper summarizes the current status of recent developments in the long-developing problem of the effects that engine emissions ( $\text{NO}_x$ ,  $\text{H}_2\text{O}$ ,  $\text{SO}_2$ , etc.) from aircraft at cruise altitudes may have on the ozone layer and on climate. This work, as was the case with three prior similar papers by The Institute for Defense Analyses (IDA) (Oliver et al., 1977, 1978; Oliver, 1979) has been sponsored by the High Altitude Pollution Program (HAPP) of the Federal Aviation Administration.

A brief history of computed aircraft effects, which have been considered in some aspects at least since 1964 (Hampson, 1964), was provided by Oliver (1979). The expected advent of SSTs cruising at 17 to 20 km in the stratosphere, a previously little-traveled region of the atmosphere which contains most of the protective ozone, provided the initial stimulus. In time, the concerns shifted to include possible (although much smaller) effects of subsonic aircraft fleets (cf. Grobecker et al., 1974; NAS, 1975). Later-identified threats to ozone, such as nitrous oxides from fertilizer, and particularly halocarbons (Molina and Rowland, 1974), have been the major focus of national investigations outside of FAA since 1975; the knowledge developed has had a significant impact on the aircraft-effects question. The complexity of all these issues, evidenced by many changes over time, and the interrelated aspects of various anthropogenic perturbations, have become abundantly clear during the course of these investigations. Clearly, further developments can be expected.

A number of issues pertinent to aircraft effects are treated in this paper. The first, which provides an input to the overall problem, relates to projected rates and the altitudes at which pollutants will be injected into the atmosphere: the rate depends on traffic levels (fuel used) and emissions per unit of fuel burned. Computed effects on ozone at arbitrary

large emission rates -- in effect, standard problems -- and how these have varied with time, are then treated, showing the strong effects of changes in the reaction rates used. Interaction with other projected changes are also noted. Other effects, based on recently published information, are discussed briefly.

## 2.0 AIRCRAFT EMISSIONS PROJECTIONS

### 2.1 INTRODUCTION

The rate at which pollutants are put into the atmosphere by aircraft at cruise altitudes depends on the product of fuel burned per unit of time and the emission index (g/kg fuel) for the pollutant species of interest. These are allocated, depending on the model being used, by: altitude (1-D); altitude and latitude (2-D); or altitude, latitude, and longitude (3-D). Generation of such detailed information is time-consuming and subject, for projections, to the vagaries of forecasting. In addition, for the important constituent  $\text{NO}_x$ , there has been a measurement uncertainty, in that probe results have been questioned, based on results of a spectroscopic technique first employed at the Arnold Engineering Development Center (AEDC) (McGergor et al., 1972) and investigated periodically since.

Only one estimate (Athens et al., 1976) of projected aircraft fleet emissions has been made in sufficient detail for modeling purposes, and this is becoming obsolete. However, periodic updates of traffic and fuel consumption projects are made, and these provide some guidance (Section 2.2). In addition, there has been development in the emission measurement area, as discussed in Section 2.3. A brief discussion of  $\text{NO}_x$  emission measurement efforts is presented in Section 2.4. A 1990 2-D base-case emission model is included in Section 2.5.

### 2.2 AIR-TRAFFIC AND FUEL-USE PROJECTIONS

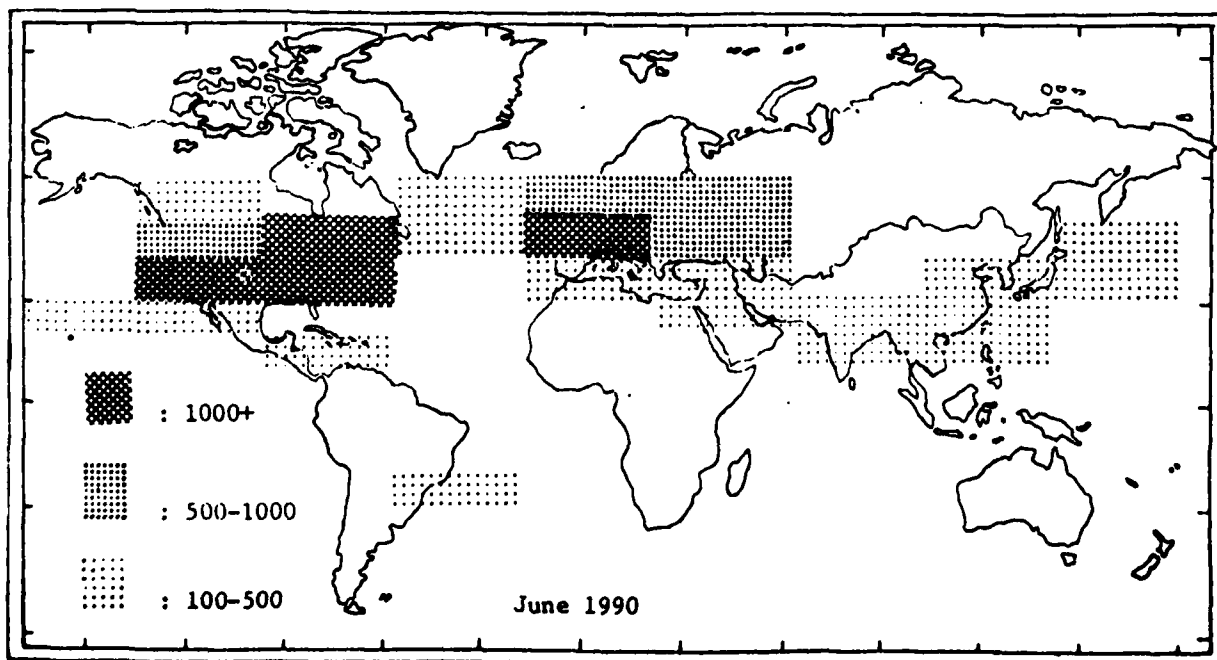
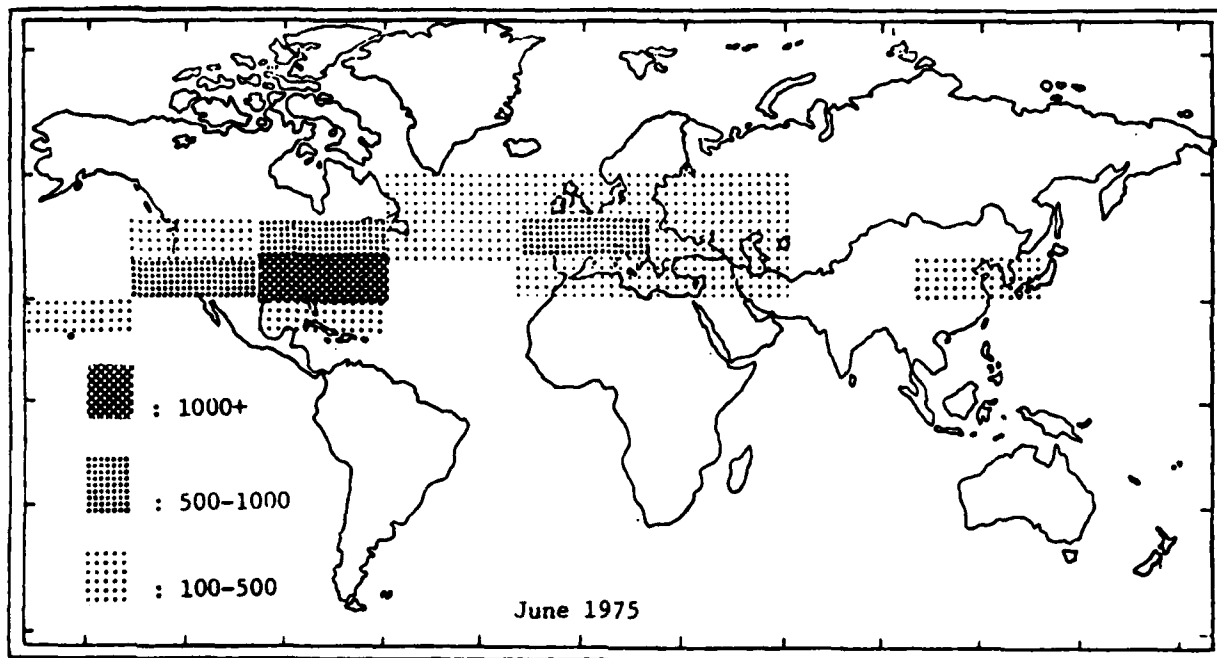
#### 2.2.1 Air Traffic--General

The geographic distribution of air traffic in 1975 and as forecast for 1990 by Pozdena\* (1976), is shown in Fig. 1. Heaviest traffic in both

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\*These were used by Athens et al. (1976) to generate emissions data.





Note: The 1990 forecast in this figure is based on the Base case.

FIGURE 1. Air Traffic Distributions (by flight hours per day) for 1975 and Projected for 1990. (Pozdena, 1976)

cases is over the Eastern United States and in the mid-latitudes in the Northern Hemisphere. The growth between 1975 and 1990 and the expected heavy European traffic should be noted. These forecasts were generated under FAA-HAPP sponsorship for the purpose of computing air traffic effects. The forecasts still provide the best breakdown available, but need examination in light of more recent events.

There are two obvious constraints on the long-term growth of air traffic. First, and probably most important, is the high cost and uncertain supply of fuel; the second is ground facilities.\* In spite of these factors, even recent projections show an expected growth in air traffic, at least over the next decade (Raphael, 1980; FAA, 1979, 1980); some fuel consumption figures are shown in Fig. 2. The upper curve in Fig. 2 (from Raphael, 1980), which does not include fuel used in the USSR or in the Peoples Republic of China, projects about a 60 percent increase between 1980 and 1990; expected passenger and freight increases are actually larger than this figure, because the curve includes projected increases in fuel efficiency. The curves labeled "high" and "base" are from Athens et al. (1976), which used Pozdena's 1976\*\* estimates of traffic as a data base; these figures represent fuel burned at altitudes above 6 km, rather than totals. Traffic restrictions at airports can slow air traffic growth and have been a matter of controversy at some heavily used airports in the U.S., such as Washington National. In fact, in the U.S. there are few large hubs where traffic constraints are not expected to influence airport capacity in the next decade (FAA, 1979). Over this period, however, the principal effect of such restrictions will be to divert traffic to nearby less-utilized airports rather than to limit total traffic significantly. Larger aircraft, with better load factors, will also be used

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\*High energy costs affect not only direct operating costs but exert indirect effects on the costs of equipment, labor, and apparently even capital, by affecting inflation rates.

\*\*The dashed line shown on Fig. 2 is the author's arbitrary projection of the estimates given by Raphael (1980). The 1990 figure (circle) in conventional units is 48 billion gal/yr., or 3.1 million 42-gal bbl/d.

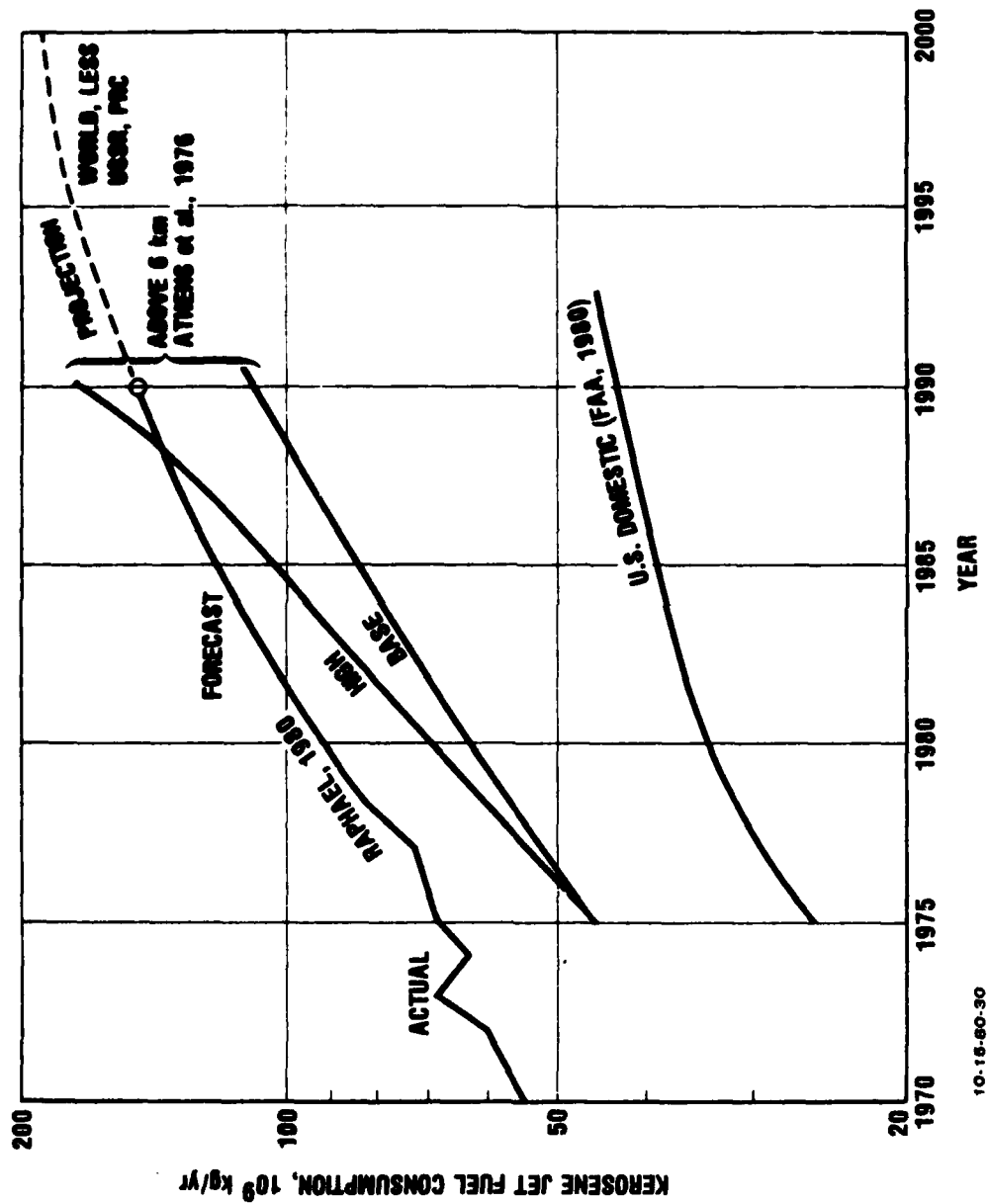


FIGURE 2. Aircraft Fleet Fuel Use.

to take up the load. (The airport capacity factor is not treated in as much detail in the 1980 FAA report as it was in the 1979 FAA report; the problem may therefore currently be viewed as less severe.)

International Air Traffic Association (IATA) projections for international travel are presented by Rek (1981) for the period to 1986. Projected growth rates are in the 6 to 8 percent range. Actual figures are presented in Table 1. The traffic distribution, by region, is expected to change only modestly, as shown by the IATA projections in Table 2.

#### 2.2.2 SST Traffic

Recent events suggest a decline rather than near-term growth in SST traffic, with Concorde flights having been cut back and the Tupolev-144 apparently being grounded.\* The possibility nevertheless exists that SSTs will, in time, fill a significant niche in the air market.\*\* Studies and technology development under NASA sponsorship continue (OTA, 1980; Powers et al., 1979; Baker, 1979). A hypothesized fleet for the year 2010, prepared for the Office of Technology Assessment (OTA, 1980), is described by Table 3. Since the source of the information presented in the table is dated January 1979, it appears that the most recent round of fuel cost increases which occurred in 1979 could not have been included in developing this fleet estimate. OTA discusses the adverse effects of high fuel costs on SSTs, but SST economic viability and environmental impact depends on a number of factors in addition to fuel cost. Impact on the ozone layer depends, for example, on cruise altitude, which increases about 3 km in going from Mach 2.0 to Mach 2.7. Bond et al. (1980) suggest that little more productivity is achieved at Mach 2.7 than at Mach 2.0; this point would surely be debated should a new SST be considered.

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\*Soviet experience with the Tu-144 (which includes two crashes) was described in Aviation Week (1978a). The last flight reported therein took place on June 1, 1978.

\*\*A news item (Maurice 1981) notes that Federal Express may lease up to four Concorde for express parcel delivery. According to the article, even at a 50 percent load factor (of a maximum of 36,000 lb), in Atlantic service, a profit margin of 14 to 15 percent could be achieved.

TABLE 1. PROJECTIONS OF INTERNATIONAL TRAVEL BY IATA (Rek, 1981)

	Actual		Forecasts					
	<u>1974</u>	<u>1980</u>	<u>1981</u>	<u>1982</u>	<u>1983</u>	<u>1984</u>	<u>1985</u>	<u>1986</u>
<u>Scheduled Passenger</u>								
19 <sup>9</sup> Passenger-km	218.4	370.9	387.7	412.6	443.9	473.8	499.1	528.8
Percent Growth Over Previous Year	--	2.5	4.5	6.4	7.5	6.7	5.3	5.9
Annual Average Growth, Percent	↔ 9.2 ↔		↔		6.1			↔
<u>Scheduled and Non- Scheduled Freight</u>								
10 <sup>6</sup> tonne-km	10,598	17,756	18,687	20,037	21,696	23,359	24,743	26,368
Percent Growth Over Previous Year	8.0	5.8	5.2	7.2	8.3	7.7	5.9	6.6
Average Annual Growth, Percent	↔ 9.0 ↔		↔		6.8			↔

TABLE 2. DISTRIBUTION OF INTERNATIONAL AIR TRAFFIC,  
1980 AND 1986 (Rek, 1981)

	<u>1980, Percent</u>	<u>1986, Percent</u>
North Atlantic	26.8	24.8
Mid and South Atlantic	5.7	5.8
Europe/Middle East	6.0	5.2
Europe/Middle East - Africa	7.0	7.0
Europe/Middle East - Far East and Southwest Pacific	15.6	16.4
North-Mid Pacific and South Pacific	8.6	9.1
North America - Central and South America	8.5	9.5
Within Europe	11.6	11.0
Other Route Areas	10.2	11.2

2.2.3 Fleet Mix Comparisons and a Recommendation for Use of  
the "Base" Case

Fleet mixes from Pozdena (1976) for the global fleet and from FAA (1980) for the U.S. domestic fleet are tabulated in Table 4. (The 1990 SST projections were thought by Pozdena to be optimistic at the time, so should not be given much weight.) Note that the expected FAA increase in total U.S. domestic jets from 1980 to 1990 is 18 percent (2428 to 2819 aircraft), whereas the 1976 global estimate over the same period for the base case showed a 27 percent increase (6934 to 8458) and the high estimate a 71 percent increase (7441 to 12,701). On this basis, as well as on the basis of the slope of the fuel consumption projections in Fig. 2, the "high case" looks increasingly unrealistic for 1990. The base case, as the only alternative, seems preferable for modeling use. Emission figures for this case are provided in Section 2.5.

TABLE 3. WORLD (LESS USSR, PRC) COMMERCIAL JET FLEET  
WITH AND WITHOUT ADVANCED SUPERSONIC TRANSPORTS -  
YEAR 2010 (OTA, 1980)

Aircraft Type <sup>a</sup>	Passenger Seats	Fleet		No. of Subsonic Aircraft Replaced by AST
		Without AST	With AST	
Short and Medium Haul				
2S	100	150	150	--
2S	130	700	700	--
2S	160	1,200	1,200	--
2W	200	2,000	2,000	--
3W	250	1,550	1,550	--
3W	290	400	400	--
Long Haul				
3W	200 LR <sup>b</sup>	150	100	50
3W	250 LR	400	200	200
4W	420 LR	750	350	400
4W	530 LR	500	400	100
4W	600 LR	300	200	100
4AST	330	-	400	-
TOTALS		8,100	7,250 Subsonic 400 Supersonic	850

<sup>a</sup>Aircraft are classified by the number of engines (2, 3, or 4) and by body (S = standard, W = wide); AST = advanced supersonic transport.

<sup>b</sup>LR = Long-range flights.

Source: OTA Working Paper, Boeing Commercial Airplane Co., 1/22/79.

TABLE 4. JET AIRCRAFT FORECASTS

	In Service of U.S. Air Carriers (FAA, 1980)				Global* (Pozdena, 1976)						
	<u>Baseline</u>				<u>Base</u>				<u>High</u>		
	1975	1980	1985	1990	1975	1980	1985	1990	1980	1985	1990
<u>Supersonic</u>	-	-	-	-	-	20	47	88	22	55	131
<u>Four-Engine</u>	627	501	344	357	1639	1856	1862	1792	1929	2203	2791
Wide Body	-	-	-	-	236	416	771	1167	469	1033	1887
Standard Body	-	-	-	-	1403	1440	1091	625	1460	1170	904
<u>Three-Engine</u>	926	1262	1370	1364	2045	3115	3530	3751	3470	4539	5916
Wide Body	-	-	-	-	260	686	1185	1878	844	1569	2971
Standard Body	-	-	-	-	1785	2429	2345	1873	2626	2970	2945
<u>Two-Engine</u>	541	665	927	1148	1805	1943	2514	2827	2020	3068	3863
Wide Body	-	-	-	-	5	35	694	1485	50	1242	2273
Standard Body	-	-	-	-	1800	1908	1820	1342	1970	1826	1590
<b>TOTALS</b>	<b>2094</b>	<b>2428</b>	<b>2641</b>	<b>2869</b>	<b>5489</b>	<b>6934</b>	<b>7953</b>	<b>8458</b>	<b>7441</b>	<b>9865</b>	<b>12,701</b>

\*Greater than 400 nmi.

It is also of interest to note that the 1980 FAA projections show a drop in four-engine aircraft between 1980 and 1990 of 29 percent, whereas the 1976 projections for the base case showed a drop of only 3 percent, and the high case an increase of 45 percent. The recent more rapid decline in four-engine jets undoubtedly reflects a more rapid phaseout of old narrow-bodied aircraft (such as the 707) than was anticipated in 1976, due obviously to increases in fuel costs.

Note that the difference in Fig. 2 between the "above 6 km" fuel figures and the total figures implies a fairly large (and somewhat questionable) fuel consumption below 6 km. New projections are needed, as noted earlier.



## 2.3 EMISSION MEASUREMENTS AND DISCUSSION

### 2.3.1 NO<sub>x</sub> Emission Indices

The accuracy of probe measurements in determining NO<sub>x</sub> (the emittant of greatest interest) in aircraft exhausts has been at issue since 1972 when workers at AEDC (McGregor et al., 1972; Davidson and Domal, 1973) reported on the results they obtained by using an optical (UV spectroscopy) technique. These results showed NO concentrations 1.5 to 5 times those found with probes. The NO measurements were taken on a YJ93GE3 engine under simulated flight conditions at altitudes ranging from 35,000 ft to 75,000 ft and at speeds ranging from Mach 1.4 to 2.6. The highest ratios of optical to probe NO occurred under maximum afterburning conditions, the ratio increasing with altitude. The measurements, or samples, were taken only 4 to 12 inches from the engine exhaust plane in a still-reacting plume (total temperature to 3100° F). Hydroxyl (OH) concentrations were also measured, and in some cases exceeded those of NO (one OH concentration measured was 776 ppm, as discussed in Section 2.3.2).

The issues raised led to a number of further investigations (for example, see Lyon et al., 1975; Few et al., 1979) which have been discussed in previous IDA summaries. The most recent development in this area is completion of work at United Technology Research Center (UTRC) (Dodge et al., 1979; Colket et al., 1980; Zabielski et al., 1980) under FAA-HAPP sponsorship. These investigators undertook a detailed study of probes and the AEDC ultraviolet spectroscopic technique; an investigation using infrared techniques (which had also been used earlier in an investigation at Wright-Patterson AFB) was pursued as a subtask by D.A. Gryvnak at Ford Aerospace and Communications (see Appendix A of Zabielski et al., 1980). The tests were not run on actual engines, but on a flat-flame burner using CH<sub>4</sub>, a research swirl burner burning C<sub>3</sub>H<sub>8</sub>, and a modified FT12 combustor using Jet A fuel. In all cases, the airstream was enriched with NO (ca. 800 ppm) to obtain concentration levels which would give suitable absorptions in the spectroscopic methods over the path lengths employed. Centerline temperatures ranging from 600 K to 1800 K, and fuel mixtures ranging from lean to rich were tested.

The UTRC work uncovered errors in the AEDC UV data-reduction techniques and also showed the great complexity of the spectroscopic technique, in that temperature and other data must be obtained as a function of position in the stream (which requires probes), and these data must be used in reducing the spectroscopic data. Theory must be used in extending measurements into regimes at which calibration is essentially impossible, and the theory itself is subject to argument. In any event, the UV and IR work both showed reasonable agreement with properly designed water-cooled probes under the conditions employed. It was found to be possible, using uncooled probes in fuel-rich conditions, to destroy up to 80 percent of the NO reaching the probe.

The UTRC work was carried out in cooperation with the AEDC group. Computer programs were corrected and substantially agreed to by both groups. The AEDC group (Few and Lowry, 1981) then reevaluated earlier results, but found that the revision did not substantially alter their earlier conclusions, where high ratios of optical/probe values for NO were found. Some of these revised results are presented in Table 5. Temperature and other data are by Few and Lowry (1981).

Few and Lowry argue that their previous results for high-velocity (supersonic), high-temperature airstreams are not substantially modified with use of the corrected program. (Both the YJ93 and the Avco-Lycoming cases above are in supersonic streams.) The criterion is probably not one of Mach number, but rather a question of whether the probe is in a still-reacting stream, which is more probable with higher-velocity streams.

Few and Lowry also report substantially revised measurements for an F101-GF-100 engine under simulated conditions of 11 km altitude and Mach 0.95. The newer UV technique gives good agreement with the probe radially outward of about 15 percent of the nozzle radius, but there are discrepancies of nearly a factor of 2 at the centerline.

Few and Lowry conclude that for subsonic, non-reacting flows, and for the mixed fan flow region of turbofan engine exhaust, the results for probe and optical techniques agree. This condition does not apply to the core region.

TABLE 5. AEDC NO<sub>x</sub> RESULTS REVISED  
(Few and Lowry, 1981)

	Power Level or Fuel/Air	Mach	Pressure or Altitudes, ft	NO <sub>x</sub> , ppmv			Ratios ( <u>New Optical</u> Probe)
				Probe	Optical Old	Optical New	
<u>YJ936E-3</u>	Military	2.6	65,000	100	323	436	4.36
(4 to 12 in. from exit plane)	Maximum After- burning	2.6	65,000	130	617	686	5.28
<u>T56 Combustor</u>	a	b	(14.7 psia)	109	170	119	1.09
(7.62 cm from nozzle exit)	a	b		125	190	129	1.03
	a	b		154	270	181	1.17
<u>Avco-Lycoming</u>	0.01	c		30	175	82	2.73
<u>Combustor</u>	0.02	c		102	605	305	3.00
(1.27 cm from nozzle)	0.03	c		190	860	617	3.25
	0.04	c		215	1220	953	4.43
	0.05	c		264	1600	1236	4.68

- a. Pure fuel, or fuel with pyridine added.  
b. Low subsonic.  
c. Slightly supersonic.

The UTRC authors point out the corrections resulting from use of a correct spectral model and other factors in using the spectroscopic technique (note the old and new T56 combustor results above), and comment on some of the problems in the measuring of NO in supersonic streams. Unless measurements are taken with great care, both the optical and probe results may be in error.

The large ratios shown in Table 5 are obviously troublesome. There seems to be no unambiguous resolution to the uncertainties implied, and continuing investigations are apparently not underway. Note, however, that errors of the magnitude indicated, if present in actual engine measurements, should have led to large increases in apparent emission index at downstream positions as the gas stream was diluted and cooled. No

such trend was evident, for example, in the 1973 J85 tests cited in Table 6. In these results the apparent  $\text{NO}_x$  emission index often dropped, instead of increasing, with dilution.

TABLE 6. J85  $\text{NO}_x$  RESULTS (German et al., 1973)\*

<u>E.I. <math>\text{NO}_x</math>, g/kg at Position, Exit Diameters Downstream</u>					
<u>M<sub>∞</sub></u>	<u>Altitude, ft</u>	<u>Power</u>	<u>Position</u>		
			<u>0.22</u>	<u>9.3</u>	<u>19.9</u>
1.6	55,000	Military	3.25	1.99	2.32
2.0	65,000	Military	4.25	3.97	3.20
1.6	55,000	Partial Afterburning	1.99	2.38	1.10
2.0	65,000	Partial Afterburning	2.15	2.20	1.78

\*Also reported in English et al., 1975.

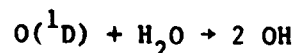
Another major investigation in this area was that reported by Lyon et al. (1975), using J85-5 and J79-5 engines under sea-level conditions at military and three afterburning power levels; the results were reviewed in a report by Oliver et al. (1978). Lyon et al. found no trend in emission index with dilution, and concluded that their "far plume" procedure provided reliable estimates of the residual emissions put into the atmosphere, even under afterburning conditions. A reexamination of the data by this author (see Appendix in Oliver et al., 1978) did suggest a possible upward trend in emission indices with increasing dilution, but the procedure was sensitive to background concentrations and instrumentation questions. The apparent trend was not noticeably different between military power and afterburning conditions, which disagrees with the AEDC observations. The increase in apparent emission index was, at most, a factor of 2 or 3, rather than 5 or more, and most noticeable at very high dilutions ( $10^4$  to 1). The analysis was thus perhaps interesting, but hardly conclusive.

To conclude this discussion, it is now agreed that probe and optical methods, when both are carried out properly, effectively provide the same  $\text{NO}_x$  results for low-velocity subsonic streams: probe data are probably satisfactory for subsonic aircraft but some possibility still exists that emission estimates may be in modest error due to probe sampling errors in the jet core. Some further investigation into high velocity (transonic and supersonic) stream-sampling techniques still may be in order, but the problems involved are complex. This author would prefer to see "simple" procedures attempted (such as analysis of gas samples taken from a diluted stream, rather than any attempts to find all the possible problems which may be inherent in more sophisticated procedures. It seems unreasonable that discrepancies of the magnitude indicated (factors of 4 or more) would not have led to obvious anomalies somewhere along the line in the test of the J85 and GE engines. Only probe data are used here.

### 2.3.2 OH Emissions and Other Emissions Affecting OH

Measurements by UV spectroscopy taken during CIAP showed surprising amounts of OH in the aircraft exhaust. While any resulting effects on the ozone problem appeared to be negligible, based on CIAP wake studies, and though the UV spectroscopic methods are somewhat suspect, the question seems worthy of brief reexamination in view of the recognized critical effect of OH concentrations on calculated results.

The question can be put in perspective by computing the OH production rate from natural processes, and by comparing them to plausible injection rates. This can be done by using the relation



and

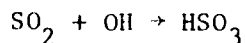
$$n_{\text{OH}} = 2k[\text{O}(^1\text{D})][\text{H}_2\text{O}]$$

where  $n_{\text{OH}}$  is in molecules  $\text{cm}^3\text{-sec}^{-1}$ ,  $k = 2.3 \times 10^{-10} \text{ cm}^3\text{sec}^{-1}$  (Hudson and Reed, 1979),  $[\text{O}(^1\text{D})] \cong 0.4$  at 17 km, or  $0.8 \text{ cm}^{-3}$  at 20 km (CIAP, Monograph 1, pp. 3-93) and  $[\text{O}(^1\text{D})] \cong 9 \times 10^{12} \text{ cm}^{-3}$  at 17 km and  $5 \times 10^{12} \text{ cm}^{-3}$  at 20 km. To first order, it follows that the production rate in this

altitude regime is about 2000 molecules of OH  $\text{cm}^{-3}$   $\text{-sec}^{-1}$ . Since "standard" ozone modeling problems tend to work with injections of the order of 1000 molecules  $\text{cm}^{-3}$   $\text{-sec}^{-1}$ , over only 1 km altitude range, it would seem plausible that an injection of OH molecules of the same order of magnitude would be required to significantly affect the chemistry, and this effect would tend to be localized vertically. Whether such OH emissions might be involved can be deduced from the OH measurements reported by Davidson and Domal (1973) and included in Table 7. These measurements were made on a J93 engine at simulated altitude and Mach number conditions. As can be seen in Table 7, OH concentrations were in fact found in some cases to approximate NO concentrations, but only under afterburning conditions.

Davidson and Domal (1973) compared their results to theoretical values under stagnation conditions. Hoshizaki developed kinetic values (Chapter 2 of Robinson et al., 1975). In some cases, agreement was reasonable, but in all cases at military power (approximately cruise), the measured values were far above the theoretical values. In any event, as an upper bound, the possibility exists that OH emissions under cruise conditions, without afterburning, may reach 10 percent of NO (molecular) emission levels. In view of the discussion above, such OH emissions would seem to be of little interest, even at standard high NO injection rates. With afterburning, however, the effect may be more significant,\* although other radicals would then need to be examined. It is possible that the level of OH to NO may rise as NO emissions are reduced. OH production might also be increased deliberately by using some afterburning.

SO<sub>2</sub> is emitted by aircraft burning sulphur-containing fuels, a nominal value, as used in CIAP, being 1 g/kg, corresponding to 0.05 percent sulphur in the fuel. SO<sub>2</sub> reacts with OH according to




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\*Note that the GE4 engine for the cancelled Boeing 2707 SST was to be operated in afterburning mode during cruise (English et al., 1975, p. 4-43).

and by further processes to give  $\text{H}_2\text{SO}_4$ . It would not be unreasonable to assume 2 OH molecules are eliminated per  $\text{SO}_2$  molecule emitted. The rate is fairly slow, but tends to reduce background OH, increasing ozone sensitivity to  $\text{NO}_x$  emissions.

TABLE 7. AEDC OH MEASUREMENTS (Davidson and Domal, 1973)

Mach	Altitude, ft	Power	$\phi^a$	<u>Observed Concentrations, ppmv</u>			
				NO(probe)	NO(UV) <sup>b</sup>	OH (UV)	OH (Theory) <sup>c</sup>
1.4	35,000	Military	0.318	51	75	5.7	0.4
1.4	35,000	A/B	0.843	--	--	776	690
2.0	55,000	Military	0.294	72	165	--	--
2.0	75,000	Military	0.290	--	--	41.7	0.6
2.0	75,000	A/B		--	--	454	396
2.6	65,000	Military	0.223	90	323(436)	22.9	0.8 2.4 <sup>d</sup>
2.6	65,000	A/B	0.560	93	617(686)	594	452,180 <sup>d</sup>

<sup>a</sup>Setting, not always confirmed by exhaust,  $f/a = \phi(0.066)$

<sup>b</sup>Original results; revised (1981) figures in parentheses

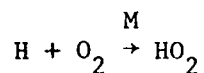
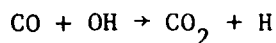
<sup>c</sup>Equilibrium at stagnation conditions (Davidson and Domal, 1973)

<sup>d</sup>Cruise at 65,000 ft at Mach 2.7 (kinetic, Hoshizaki)

Unburned hydrocarbons, CO, soot, and lubricating oil are also emitted by jet engines in cruise, but in small amounts, as combustion efficiencies of 99.9 percent are expected (lower for afterburning or ductburning engines). The following values are given in English et al. (1975), in g/kg of fuel:

Soot (as carbon)	0.1
Lubricating oil	0.1
Carbon monoxide	3.5 Concorde; 4 subsonics
Total hydrocarbons	0.2 Concorde; 0.1 subsonics

These ingredients react with OH, as for example, through



but may not destroy odd oxygen (OH, O, O<sub>3</sub>) in the process.

No model results are available on possible effects of OH, SO<sub>2</sub>, and other emissions. These are secondary effects, which may be worth considering if computed effects of NO<sub>x</sub> and H<sub>2</sub>O reach some form of stable consensus.

#### 2.4 EMISSION REDUCTION EFFORTS

Efforts to develop emission standards and the engines to meet them have been underway for about a decade, but have been directed mainly to emissions around airports.\* (See, e.g., Kittredge, 1977; Jones, 1977; Petrash et al., 1979; Sundararaman, 1978, 1979; Yamartino et al., 1980.) A set of standards was promulgated on July 17, 1973 which were known as the "1979" standards; a revised proposal was issued on May 24, 1978 which became known as the "1981" standards, and a 2-year postponement was announced for these "as an interim measure" on December 31, 1980. (EPA, 1973, 1978, 1980.) The NO<sub>x</sub> standards are now scheduled to apply 3 yrs after the CO and HC standards, or January 1, 1986. Whether these standards will be imposed or not is obviously unclear. Even if they are imposed, the effect on cruise emissions is difficult or impossible to estimate. This difficulty is partly due to the fact that proposed NO<sub>x</sub> and other standards are based on the "EPA parameter" or EPAP (which sums emissions over a specified landing and take-off cycle, and divides by the rated thrust), and partly because different standards, depending on thrust, apply to different engines. Also, newly manufactured engines are distinguished from newly certified engines, etc. Higher pressure ratio (> 25) engines (for higher fuel efficiency) also are permitted to have higher emission indices for NO<sub>x</sub> than are lower-pressure ratio engines. Industry spokesmen consider these gaseous emission standards to be "unnecessarily strict and largely unattainable" (Aviation Week, 1978).

\*Smoke standards and fuel-dumping procedures have been in place for a longer period.



As suggested, the above complications and uncertainties make impractical any attempt at estimating effects at cruise of possible emission reductions at ground level. Most emphasis in such work has been on HC and CO reductions at ground level, rather than on  $\text{NO}_x$ . It is worth noting, however, that engines have been developed, such as the CFM56 (Lyon et al., 1979), which have the goals of being both clean-burning and fuel-efficient, and which may provide lower  $\text{NO}_x$  emissions engines. However, fuel efficiency considerations, which lead to higher-pressure-ratio engines and higher  $\text{NO}_x$  values may counteract gains achieved in combustor technology. All in all, reductions at cruise below these derived from the 1976 computations (Athens et al., 1976) seem problematical.

## 2.5 THE 1990 BASE CASE

It has been noted that the 1990 "high" case (Athens et al., 1976; Oliver et al., 1977) used in previous modeling efforts now appears unrealistic, that emission measurements (at least for subsonics) are probably satisfactory, and that emission-reduction efforts may have little impact by 1990. Accordingly, the 1990 "base" case developed by Pozdena (1976) and Athens et al. (1976), and as modified by this author (as was done for the "high" case in 1977) is given here.

Fuel use reported by Athens et al. (1976) is given in Table 8; total fleet consumption as well as the SST portion is given. As was discussed in Oliver et al. (1977), the SST altitude distribution of Athens et al. appears to be at variance with prior estimates developed in CIAP. A simple correction was suggested so that these data could be used for estimates of SST emissions; the procedure involved adjusting the tabulated fuel flows (and emissions) at 15 to 16 km and 17 to 18 km each upward by 1 km and lowering the 17 to 18 km figures by 2 km.  $\text{NO}_x$  emissions are given in Table 9. In preparing Table 9, the emission indices used for  $\text{NO}_x$  for CF-6 aircraft were adjusted upward. Athens et al. (1976) quoted  $\text{NO}_x$  figures for these aircraft which were severalfold lower than for JT9-D aircraft, whereas in CIAP (English et al., 1975) the two were given the same emission index at cruise. Accordingly, as was done in Oliver et al. (1977), the  $\text{NO}_x$  emissions were adjusted by changing all "CF-6" aircraft to have the same emission indices as for "JT9-D" aircraft.

TABLE 8. FLEET AND SST FUEL USAGE FOR 1990 BASE CASE (Athens et al., 1976)

1990 WORLDWIDE AIRCRAFT FUEL USAGE, BASE ESTIMATE; AS REPORTED

Alt (km)	6-9	9-10	10-11	11-12	12-13	13-14	14-15	15-16	16-17	17-18	18-19
N. Pole	1.24E 00	1.22E 00	5.32E 00	6.32E 00	6.64E 07	4.31E 07	1.27E 07	8.46E 07	7.27E 07	5.94E 07	4.95E 07
50 to 60	0.17E 00	1.02E 00	3.62E 00	4.91E 00	4.61E 00	1.50E 00	6.55E 07	3.30E 00	2.01E 00	2.30E 00	1.29E 00
40 to 50	2.20E 00	2.00E 00	6.71E 00	1.34E 10	2.00E 00	2.00E 00	6.50E 07	2.20E 00	1.00E 00	1.40E 00	0.10E 07
30 to 40	2.27E 00	3.00E 00	6.19E 00	7.51E 00	1.00E 00	1.33E 00	5.20E 07	0.01E 07	7.00E 07	4.00E 07	2.97E 07
20 to 30	7.64E 00	9.70E 00	2.44E 00	2.00E 00	4.30E 00	6.40E 07	3.43E 07	6.00E 07	5.74E 07	2.76E 07	1.59E 07
10 to 20	3.17E 00	3.00E 00	9.60E 00	1.20E 00	1.02E 00	2.11E 07	4.07E 06	1.70E 07	1.45E 07	1.11E 07	5.00E 06
0 to 10	1.42E 00	1.72E 00	5.44E 00	5.32E 00	7.33E 07	9.04E 06	0.0	1.32E 07	1.17E 07	9.00E 06	5.57E 06
-10 to 0	0.13E 00	1.07E 00	4.64E 00	4.77E 00	4.97E 07	7.29E 06	0.0	1.37E 07	1.22E 07	1.03E 07	5.60E 06
-20 to -10	0.91E 07	1.20E 00	4.29E 00	5.04E 00	6.02E 07	1.35E 07	4.07E 06	7.14E 06	5.40E 06	3.77E 06	2.57E 06
-30 to -20	1.16E 00	1.47E 00	3.64E 00	3.93E 00	5.01E 07	2.06E 06	0.0	4.57E 06	4.00E 06	3.44E 06	2.31E 06
-40 to -30	1.20E 00	1.00E 00	2.61E 00	2.77E 00	6.32E 07	3.41E 06	1.59E 06	1.46E 06	9.05E 06	5.34E 06	2.02E 05
-50 to -40	0.00E 00	1.10E 07	1.64E 07	2.01E 07	4.07E 06	1.24E 06	0.0	0.0	0.0	0.0	0.0
-60 to -50	1.32E 06	1.10E 06	1.05E 06	4.04E 06	6.90E 06	0.0	0.0	0.0	0.0	0.0	0.0
S. Pole	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

TOTAL OF ALL FUEL = 1.00776E 11 (kilograms/year)

1990 WORLDWIDE SST FUEL USAGE, BASE ESTIMATE; AS REPORTED

Alt (km)	6-9	9-10	10-11	11-12	12-13	13-14	14-15	15-16	16-17	17-18	18-19
N. Pole	2.29E 07	2.10E 07	1.81E 07	1.00E 07	1.54E 07	1.40E 07	1.27E 07	8.46E 07	7.27E 07	5.94E 07	4.95E 07
50 to 60	1.17E 00	1.02E 00	6.79E 07	8.14E 07	7.40E 07	6.70E 07	6.55E 07	3.30E 00	2.01E 00	2.30E 00	1.29E 00
40 to 50	1.07E 00	9.50E 07	8.19E 07	7.50E 07	6.97E 07	6.31E 07	6.50E 07	2.20E 00	1.00E 00	1.40E 00	0.10E 07
30 to 40	0.99E 07	8.21E 07	7.65E 07	6.53E 07	6.02E 07	5.40E 07	5.20E 07	0.01E 07	7.00E 07	4.00E 07	2.97E 07
20 to 30	6.62E 07	4.17E 07	3.60E 07	3.32E 07	3.96E 07	2.77E 07	3.43E 07	6.00E 07	5.74E 07	2.76E 07	1.59E 07
10 to 20	0.67E 06	7.72E 06	6.64E 06	6.15E 06	5.64E 06	5.13E 06	4.47E 06	1.70E 07	1.45E 07	1.11E 07	5.00E 06
0 to 10	2.68E 05	0.0	0.0	0.0	0.0	0.0	0.0	1.32E 07	1.17E 07	9.00E 06	5.57E 06
-10 to 0	2.40E 05	0.0	0.0	0.0	0.0	0.0	0.0	1.37E 07	1.22E 07	1.03E 07	5.60E 06
-20 to -10	7.33E 06	6.73E 06	5.81E 06	5.37E 06	4.94E 06	4.47E 06	4.07E 06	7.14E 06	5.40E 06	3.77E 06	2.57E 06
-30 to -20	0.94E 06	0.0	0.0	0.0	0.0	0.0	0.0	4.57E 06	4.00E 06	3.44E 06	2.31E 06
-40 to -30	2.06E 06	2.63E 06	2.45E 06	2.10E 06	1.93E 06	1.75E 06	1.59E 06	1.46E 06	9.05E 06	5.34E 06	2.02E 05
-50 to -40	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-60 to -50	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
S. Pole	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

TABLE 9. 1990 WORLDWIDE AIRCRAFT NO<sub>x</sub> EMISSIONS (as NO<sub>2</sub>) kg/yr ADJUSTED<sup>1,2</sup>

BASE CASE

Latitude	6-8	8-9	9-10	10-11	11-12	12-13	13-14	14-15	15-16	16-17	17-18	18-19	TOTALS
N 60+	2.11E6	1.91E6	9.03E6	8.23E6	9.20E6	8.29E5	6.31E5	2.54E5	1.07E6	1.61E6	1.31E6	8.89E5	3.707E7
50+	1.47E7	8.02E6	5.92E7	6.60E7	5.11E7	5.14E6	2.32E6	1.31E6	4.13E6	6.43E6	5.06E6	2.32E6	2.257E8
40+	4.82E7	5.51E7	1.14E8	1.76E8	1.02E8	1.57E7	2.92E6	1.30E6	2.59E6	4.33E6	3.39E6	1.47E6	5.27E8
30+	4.94E7	5.89E7	1.06E8	1.98E8	1.10E8	1.90E7	1.92E6	1.06E6	8.06E5	1.67E6	1.27E6	5.34E5	5.486E8
20+	1.63E7	1.76E7	4.22E7	6.36E7	4.33E7	5.41E6	9.51E5	7.24E5	4.95E5	1.15E6	1.03E6	2.87E5	1.930E8
10+	6.80E6	7.21E6	1.63E7	2.62E7	1.78E7	2.23E6	2.88E5	9.33E4	2.00E5	3.32E5	2.61E5	1.06E5	7.780E7
0+	2.97E6	3.17E6	9.31E6	1.12E7	8.44E6	7.69E5	1.07E5	0	1.79E5	2.51E5	2.12E5	1.00E5	3.671E7
0-	2.11E6	2.40E6	7.62E6	8.73E6	6.82E6	5.72E5	8.56E4	0	1.86E5	2.61E5	2.19E5	1.02E5	2.917E7
10-	1.74E6	2.03E6	7.21E6	9.65E6	7.26E6	7.01E5	1.95E5	8.14E4	6.78E4	1.36E5	9.73E4	4.67E4	2.921E7
20-	2.33E6	2.55E6	6.01E6	8.73E6	5.49E6	5.95E5	3.26E4	0	6.18E4	8.68E4	7.31E4	4.15E4	2.600E7
30-	2.56E6	2.97E6	4.23E6	7.54E6	3.93E6	7.77E5	5.38E4	3.08E4	9.61E3	2.78E4	1.77E4	3.63E3	2.215E7
40-	1.51E5	1.95E5	2.30E5	5.25E5	2.84E5	5.89E4	1.07E1	0	0	0	0	0	1.444E6
50-	3.36E4	2.38E4	1.87E4	1.58E4	6.73E3	8.93E2	0	0	0	0	0	0	9.652E4
Sea*	0	0	0	0	0	0	0	0	0	0	0	0	0
TOTALS	1.204E3	1.621E8	3.814E8	5.844E8	3.656E8	5.178E7	9.504E6	4.854E6	9.795E6	1.628E7	1.294E7	5.899E6	1.755E9

Reference: ADL (1976), p. E-16.

<sup>1</sup>To make all "CF-6" aircraft have same emission indices as "JT-9D" aircraft.

<sup>2</sup>To distribute SST emissions in the 15-18 km band more closely to prior estimates.

See Oliver et al. (1977)

## 2.6 OTHER SOURCES OF NO<sub>x</sub>

Two papers have been completed at IDA under FAA/HAPP sponsorship which provide background information that may be needed in computing aircraft effects. The first of these (Kowalczyk and Bauer, 1981) is a review of lightning-generated NO<sub>x</sub>, and provides annual NO<sub>x</sub> emissions for lightning in a 2-D format. The second (Bauer, 1981) is a comprehensive review of natural and man-made sources of NO<sub>x</sub>, including aircraft emissions, again in a 2-D format. An abbreviated summary of these sources is given in Table 10.

TABLE 10. ATMOSPHERIC SOURCES OF ODD NITROGEN

<u>Source</u>	<u>Injection Rate (Tg/N/yr)</u>	<u>Mean Injection Height (km)</u>	<u>Latitudinal Range of Injection</u>
<u>Aircraft</u>	0.15 (1975) 0.53 (1990)	6-16 km	Northern Hemisphere, mid/high latitudes
<u>Fossil Fuel Combustion</u>	19.0 (1975) 27.0 (1990)	Ground Level Ground Level	Northern Hemisphere, mid-latitude
<u>Biomass Burning</u>			
Forest Fires	1.7	1-2 km	Tropics
Other	3.3 (1975) 3.8 (1990)	Ground Level Ground Level	
<u>Lightning</u>	5.7	7-12 km and Lower	Tropics, land only
<u>Transport from Stratosphere</u>	0.5*	Tropopause	
<u>Cosmic Rays</u>	0.06	Upper Troposphere	
<u>Exhalation from Soils</u>	~ 10.0	Ground Level	
<u>NH<sub>3</sub> Decomposition</u>	< 8.0	Throughout Troposphere	

\*Total odd nitrogen of which 5-20 percent is NO<sub>x</sub>.

### 3.0 DEVELOPMENTS IN COMPUTATIONS OF AIRCRAFT EFFECTS ON OZONE

#### 3.1 HISTORY OF COMPUTED $\text{NO}_x$ EFFECTS (1-D)

The effects that aircraft emissions may have on the ozone column are now recognized to involve a number of subtle features, including thermal effects due to water emissions and other composition changes ( $\text{NO}_2$ ,  $\text{O}_3$ , particulates) and also other factors such as the level of  $\text{ClO}_x$  in the reference atmosphere used. There are also time-dependent factors; atmospheric response lags a stratospheric forcing function by some number of years [3 to 6 or more to each 60 percent of response (Oliver et al., 1977)] depending on injection altitude and model  $K_z$ . Furthermore, the initial response to  $\text{NO}_x$  injections can apparently be opposite to the final response for "step-wise" injections below about 15 km (see Oliver et al., 1977). These sophistications have generally received less emphasis than the straightforward  $\text{NO}_x$  question, which depends on how well atmospheric chemistry is understood, as well as on how the model simulates atmospheric motions. This understanding has changed dramatically over the last decade. The point is illustrated by the time-history depicted in Fig. 3, which calculations were reported by Lawrence Livermore National Laboratory (LLNL) (1981).

While any breakdown into periods in Fig. 3 is somewhat arbitrary, it can be argued that there have been three periods involved. The first of these is the 1970 to 1975 period during which the  $\text{NO}_x$  catalytic cycle was the principal ozone-destroying mechanism recognized,\* and during which the massive CIAP study was pursued. In the second of these, 1975 to 1979, the role of chlorine in atmospheric chemistry was studied in recognition of

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\*It was during an earlier period (ca. 1964-1970) that the SST-ozone issue was first raised. During that period,  $\text{H}_2\text{O}$  emissions were thought to reduce ozone, due to a then-exaggerated view of the importance of  $\text{HO}_x$  cycles. This period was reviewed in Oliver (1979).

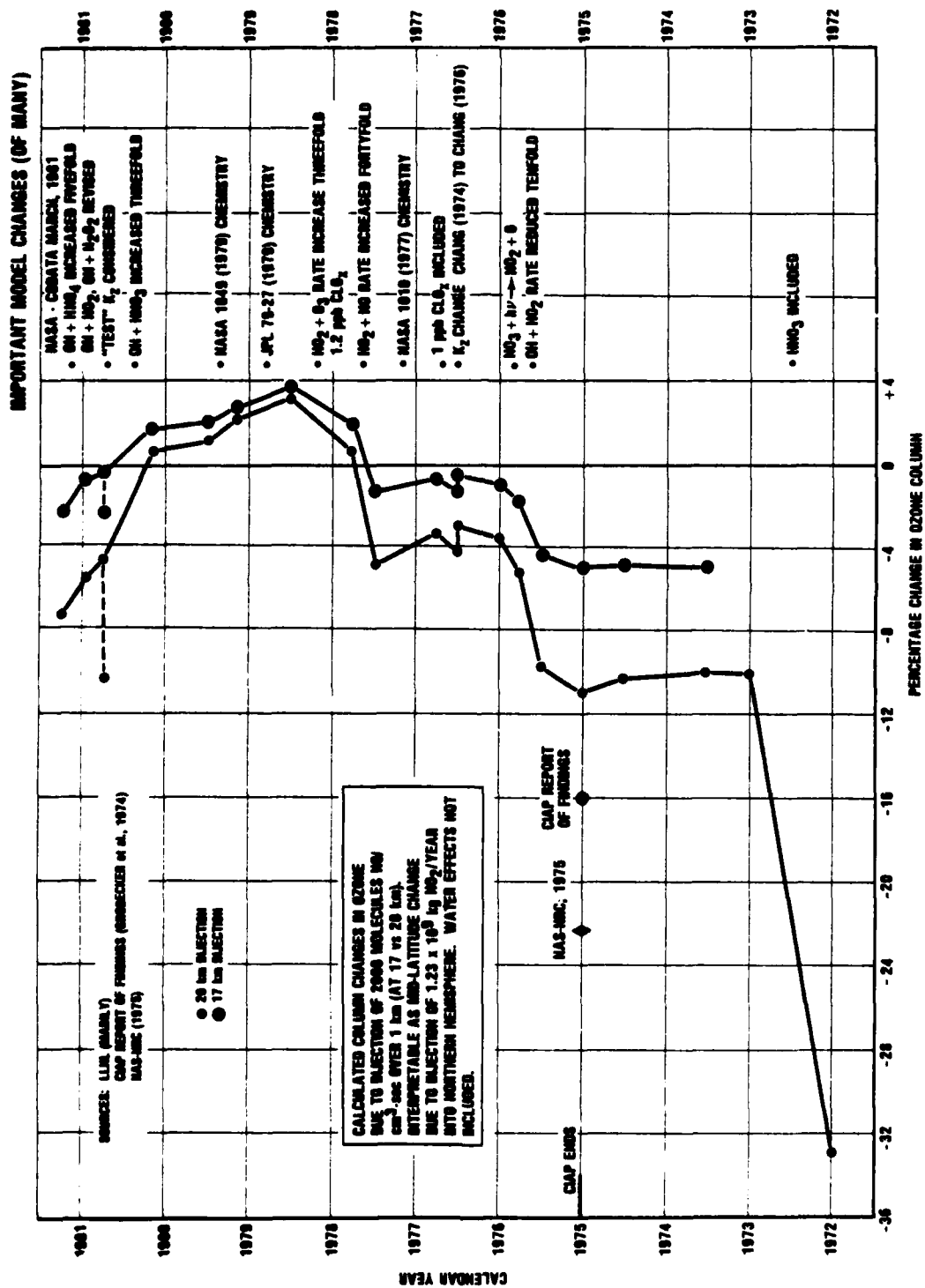


FIGURE 3. History of Computed  $\text{NO}_x$  Effects on Ozone.

the halocarbon issue (Molina and Rowland, 1974; NAS, 1976; NAS, 1979) and in which large changes were made in accepted values of reaction rates involving  $\text{HO}_2$  (with  $\text{OH}$ ,  $\text{O}_3$ ,  $\text{NO}$ ). Perhaps the most debatable of these changes has been that of the  $\text{OH} + \text{HO}_2$  rate, which was changed early in the halocarbon studies from  $2 \times 10^{-10} \text{ cm}^3/\text{sec}$ , as used in CIAP to  $2 \times 10^{-11} \text{ cm}^3/\text{sec}$ , a change which increased halocarbon effects and decreased (by about twofold)  $\text{NO}_x$  effects. The accepted rate has since been changed at least twice, first to  $4 \times 10^{-11}$  and most recently to  $8 \times 10^{-11}$  [World Meteorological Organization (WMO), 1981] with a factor of 3 uncertainty (which factor includes the nearly tenfold uncertainty range recommended in the CIAP efforts). The third period, 1979 to the present, has been dominated by revisions in the rates of  $\text{OH}$  reactions with  $\text{HNO}_3$  and  $\text{HO}_2\text{NO}_2$  (and  $\text{HO}_2$  as noted above), as well as other reactions of less impact, such as  $\text{OH}$  with  $\text{H}_2\text{O}_2$ . Considering uncertainties associated with  $K_z$  representations, these recent changes have, for practical purposes, brought the computed  $\text{NO}_x$  effects on ozone back in the general region found during CIAP at the end of 1974.

The recent revisions in the rates of reactions destroying  $\text{OH}$  (with  $\text{HNO}_3$ , etc.) have largely removed some of the questions raised by a number of authors who saw evidence that computed  $\text{OH}$  concentrations were too large in the lower stratosphere (below 30 km) in 1978-era models, a region for which no measurements exist. For example, the observed  $\text{HNO}_3/\text{NO}_2$  ratio was questioned as to compatibility with the high  $\text{OH}$  concentrations calculated (see, e.g., Hudson, 1977). Particulate formation rates from  $\text{SO}_2$  were also argued to indicate lower  $\text{OH}$  concentration (Turco et al., 1980; Whitten et al., 1981a). However, the error bars on measurements precluded firm statements, and column  $\text{OH}$  measurements (Burnett and Burnett, 1979, 1981) suggested plausibility (Hudson and Reed, 1979, p. 171), although the  $\text{OH}$  column total is not very sensitive to  $\text{OH}$  concentration uncertainties in the lower stratosphere. Recent changes in kinetics should reduce computed  $\text{HNO}_3$ ,  $\text{HO}_2\text{NO}_2$ , and  $\text{H}_2\text{O}_2$  concentrations. These changes also provide a partial explanation for failure to observe  $\text{HO}_2\text{NO}_2$ ; current model results suggest 1 ppbv rather than 1.5 ppbv  $\text{HO}_2\text{NO}_2$  (Luther, 1980), whereas 0.4 ppbv has been set as an upper bound on its presence (Murcray et al. 1979).



The most recent available calculations for  $\text{NO}_x$  effects on the ozone column (Lawrence Livermore National Laboratory, 1981) are presented in Table 11.

TABLE 11. CHANGE IN TOTAL OZONE AT STEADY STATE DUE TO INJECTIONS OF  $\text{NO}_x$  (June 1981)\*

<u>Injection Rate</u> Molecules $\text{NO sec}^{-1}\text{-cm}^{-3}$ over 1 km	<u>Ozone Change, %</u>	
	<u>Injection Altitude</u>	
	<u>17 km</u>	<u>20 km</u>
1000	-0.9	-3.4
2000	-2.2	-7.1

\*Source LLNL (1981). Chang 1976  $K_z$ .

These values now approach the values reported at the end of the CIAP program. Thus, in Grobecker et al. (1974, p. B-17), the corresponding LLNL values are -2.77 percent at 17 km and -6.29 percent at 20 km at the 1000-molecule rate and about -5 percent at 17 km and -10 percent at the 2000-molecule rate. A different  $K_z$  profile was in use at that time. Note that the "test  $K_z$ " results give larger effects than the standard 1976  $K_z$  profile (Fig. 3).

Another important change resulting from most recent revisions in reaction rates is that effects on ozone are once again computed to be roughly linear with injection rate, as was the case in CIAP (Grobecker et al., 1974). This model result differs radically from 1978-era results, in which an initial enhancement of ozone was followed by a flattening and eventually a depletion when very large injection rates were used (see Oliver, 1979).

### 3.2 OTHER COMPUTED EFFECTS (1-D)

Recent changes in reaction rates have again made prior calculations obsolete. However, some up-to-date results are available and certain important inferences can be drawn from other earlier calculations.

LLNL (1981) gives the results listed in Table 12 for steady-state effects, using their 1-D model, with the Chang 1976  $K_z$  profile, and the NASA-CODATA Chemistry (of March, 1981).

TABLE 12. OZONE PERTURBATION RESULTS AT STEADY STATE

<u>Perturbation</u>	<u><math>\Delta O_3/O_3</math></u>
Chlorofluorocarbons (CFC) ( $CFCl_3$ , $CF_2Cl_2$ , $CH_3CCl_3$ at current rates; 6.7 ppb $ClO_x$ at steady state)	-5.0%*
$N_2O$ doubled	-12.4%*
$N_2O$ doubled at CFC steady state	-12.9%*
$CO_2$ doubled	+5.8%**

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\*Source; LLNL (1981).

\*\*Penner and Luther (1981).

The CFC results imply a much-reduced threat to the ozone layer from this source; as recently as mid-1979 the corresponding result reached -19.3 percent. A doubling of  $N_2O$  was reported to result in a -2.1 percent change in ozone by the LLNL model (Hudson and Reed, 1979, p. 344), although other models showed up to 3.5 percent increases.

The most recent  $N_2O$  doubling result raises anew the fertilizer  $N_2O$ \* issue, which was first raised about 1974. Various estimates of the rate at which  $N_2O$  is released by soils, the oceans, etc., have been made, implying different rates of expected  $N_2O$  buildup. A recent careful examination of  $N_2O$  measurements, however, shows that  $N_2O$  is building up in

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\* $N_2O$  is also produced by automobile exhaust catalytic systems and in power plants.

the atmosphere at about 0.2 percent per year, with an expected increase of 5 to 7 percent by the year 2000 (Weiss, 1981). A much faster buildup might occur should manmade sources build up exponentially, and natural sinks not increase. For example, if the January 1978 figure (Weiss, 1981) of  $11 \times 10^{10}$  mol  $N_2O$ /yr as an anthropogenic source is used, along with a near infinite life for  $N_2O$ , a 4 percent growth rate, and a 2200 Mt inventory, the doubling time would be 75 years. At 2 percent, the doubling time would be 117 years.

An examination of the range of expected ozone changes, both in the column and at 40 km (3 mbar) has been conducted by LLNL (1981) using current chemistry for the interaction of CFM,  $CO_2$ , and aircraft emissions to about 1990. The interactions substantially modify expected changes, e.g., from CFM alone. The range in net effect on the column varies from about +2 percent to -3 percent in 1990. At 40 mbar, the trend (from 1970) is entirely negative, with uncertainties changing the magnitude of the effect from about -5 percent to about -7 percent.

Current computed results due to changes in water vapor,  $CO_2$ , various interactions of  $NO_x$  and halocarbons, and for subsonic aircraft emissions, are not available. Earlier results, however, show the complex interactions which may occur over time. Thus, Penner and Luther (1981), allowing for temperature feedback and hydrostatic adjustments, reported a doubling of  $CO_2$ , a 5.8 percent increase in ozone (with a 2 K surface temperature increase). This increase is due to cooling of the stratosphere and reduction in the rate of certain thermal ozone destruction reactions. A doubling of water vapor (apparently holding surface temperature fixed) led to a 1.9 percent decrease in ozone.

A number of other minor species changes have been reported to be occurring which will affect stratospheric composition and thus ozone columns. Stratospheric water vapor has been found to change with time (Pollock et al., 1980), perhaps due to changes in tropical tropopause temperatures induced by changes in solar output (Gage and Reed, 1981). CO concentrations have reportedly been increasing (Dianov-Klokov et al., 1978, Dianov-Klokev and Yurganov, 1981), about 2 percent per year since 1952 in winter only in the

north temperature zone, with an increase of 40 to 50 percent since 1952.  $\text{CH}_4$  has been found to be increasing at  $1.9 \pm 0.5$  percent per year (Rasmussen and Khalil, 1981). Increases in carbon dioxide and halocarbons of many types are well documented.  $\text{NO}_x$  sources in the troposphere, and changes therein are still being evaluated for tropospheric ozone impact (Liu et al., 1980; Noxon, 1981). Climatic effects and changes are also noted by Lacis et al. (1981), see Section 4.5. A number of ingredients not now included in atmospheric ozone modeling such as PAN (Singh and Hanst, 1981), OCS,  $\text{CS}_2$  (Wine et al., 1981), etc., may be significant in the computation of effects due to subsonic aircraft. All of these should be examined in detail if changes in ozone column are to be confidently projected.

It is of interest to note that the main hypothesized "threats" to the stratospheric ozone layer, SST  $\text{NO}_x$ ,  $\text{N}_2\text{O}$  from fertilizer and combustion, and halocarbons all deplete ozone by current models. SST  $\text{H}_2\text{O}$  may also, but this issue is more complex. It is also important to note that the threat which is of most current significance -- that due to halocarbons -- is considerably reduced in magnitude from that estimated a year or two ago. Of course, had emissions of chlorofluorocarbons continued to grow, as they had been, the "threat" would not be as well contained.

### 3.3 2-D MODEL RESULTS

The aircraft effects problem has been examined most recently in 2-D model studies by Whitten et al. (1981, 1981a) at NASA-Ames. The more recent of these two publications (1981a) which uses chemistry from Hudson and Reed (1979) updated by the changes shown in Table 13 (which table also shows current NASA CODATA recommendations), gives results, where comparison can be made, similar to those with CODATA chemistry, and are thus of current interest.\* In particular, the "new" chemistry reduces computed OH concentrations by a factor of about 2 below 25 km (which Whitten et al. point out may still be insufficient).

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\*Whitten et al. (1981a) give 10 percent average depletion of ozone for a doubling of  $\text{N}_2\text{O}$  vs. 12.4 percent reported earlier herein from LINL in their 1-D model. The chemistry set does not include HOCl nor (apparently)  $\text{HO}_2\text{NO}_2$ . The model has 2.1 ppbv of  $\text{Cl}_x$  at 60 km, which they argue to be consistent with projection to 1990, based on references they cite. Other details are provided in the two papers referenced.

TABLE 13. HO<sub>x</sub> REACTION RATE COEFFICIENTS  
(Molecules cm<sup>-3</sup> sec<sup>-1</sup>)

<u>Reaction</u>	<u>Old Value<sup>a</sup></u>	<u>New Value<sup>a</sup></u>	<u>NASA CODATA</u>
OH + HO <sub>2</sub> → H <sub>2</sub> O + O <sub>2</sub>	4.0 × 10 <sup>-11</sup>		8 × 10 <sup>-11</sup>
OH + H <sub>2</sub> O <sub>2</sub> → H <sub>2</sub> O + HO <sub>2</sub>	3.8 × 10 <sup>-13</sup>	2.5 × 10 <sup>-12</sup> exp(-126/T)	2.9 × 10 <sup>-12</sup> exp(-160/T)
OH + HNO <sub>3</sub> → H <sub>2</sub> O + NO <sub>3</sub>	8.5 × 10 <sup>-14</sup>	1.52 × 10 <sup>-14</sup> exp(649/T)	1.5 × 10 <sup>-14</sup> exp(-650/T)
HO <sub>2</sub> + HO <sub>2</sub> → H <sub>2</sub> O <sub>2</sub> + O <sub>2</sub>	2.5 × 10 <sup>-12</sup>	4.6 × 10 <sup>-14</sup> exp(1200/T)	3 × 10 <sup>-12</sup> <sup>c</sup>

a. Source: "old" and "new" values are from Whitten et al. (1981).

b. WMO (1981) draft.

c. This value is about one-third that given in the "new" value evaluated at 220 K.

Whitten et al. (1981a) postulate an SST fleet numbering 250 at 20 km altitude. Each aircraft operates 7.25 hrs/day, burning fuel at 37,800 kg/hr. The NO<sub>x</sub> emission index, assuming advanced combustion technology, is taken as 6 g (as NO<sub>2</sub>)/kg fuel; the H<sub>2</sub>O emission index is 1.3 g/mg. Corresponding total annual emissions are 1.5 × 10<sup>8</sup> kg NO<sub>x</sub> (as NO<sub>2</sub>) and 3.25 × 10<sup>10</sup> kg H<sub>2</sub>O. The route structure assumed is based on a study of long-range (> 2000 nmi), over-water routes flown by the Boeing 747 fleet, using an April 1977 official Airline Guide (worldwide edition). Almost all of the traffic was in the 30 to 60°N region.

The NASA-Ames 2-D model extends from the surface to 60 km and from 80°S to 80°N, with 5° latitudinal grid spacings. Vertical resolution is 2.5 km.

The steady-state changes in ozone resulting from the NO<sub>x</sub> and H<sub>2</sub>O injections are shown separately in Figs. 4 and 5. The "new" chemistry

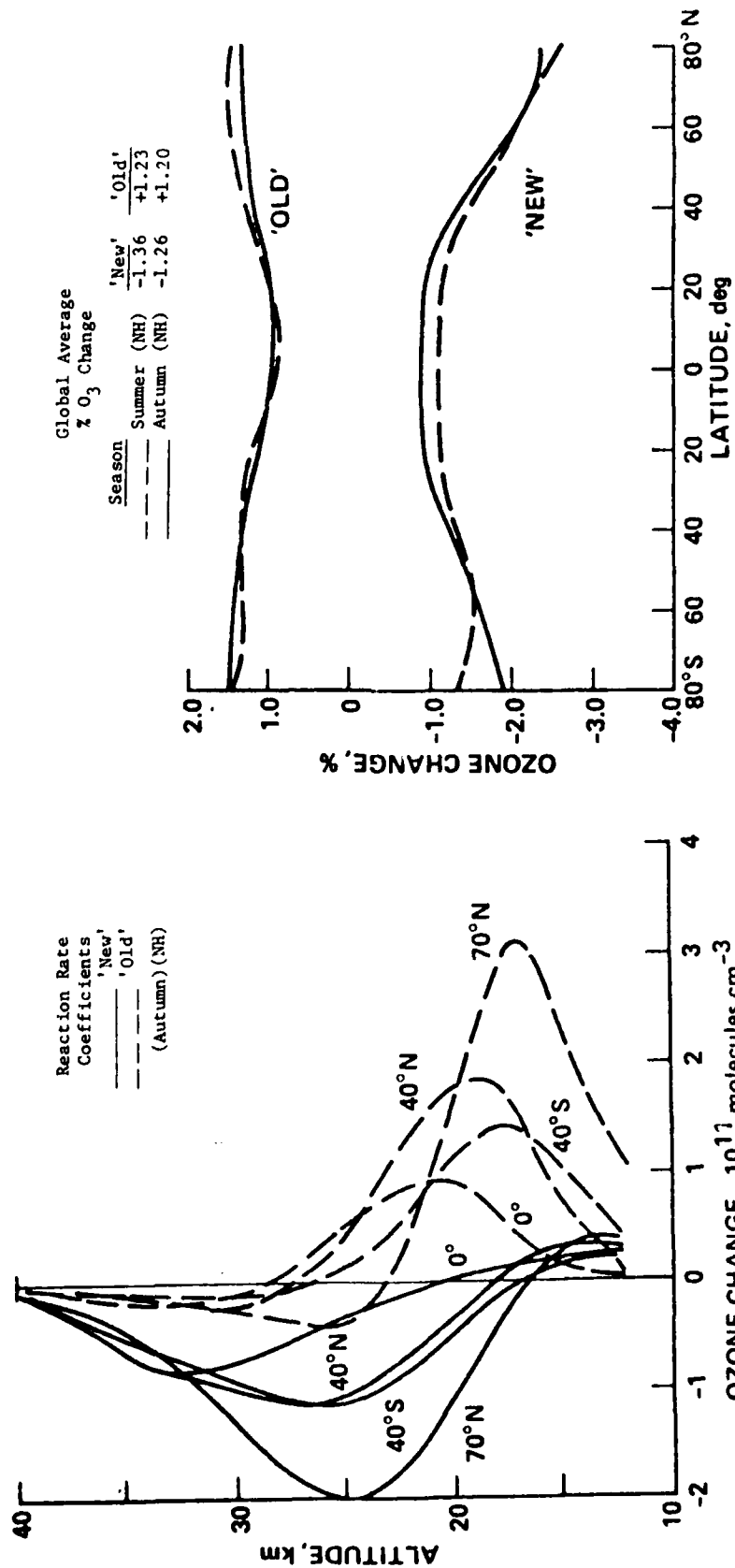


FIGURE 4. 2-D Ozone Change Results Reported by Whitten et al. (1981a) Using "Old" and "New" Chemistry Due to NO<sub>x</sub> Emissions from Assumed SST Fleet Numbering 250 and Flying at 20 km Altitude ( $1.5 \times 10^8$  kg NO<sub>x</sub> (as NO<sub>1</sub>)/yr 30 to 60°N. (Whitten et al., 1981a)

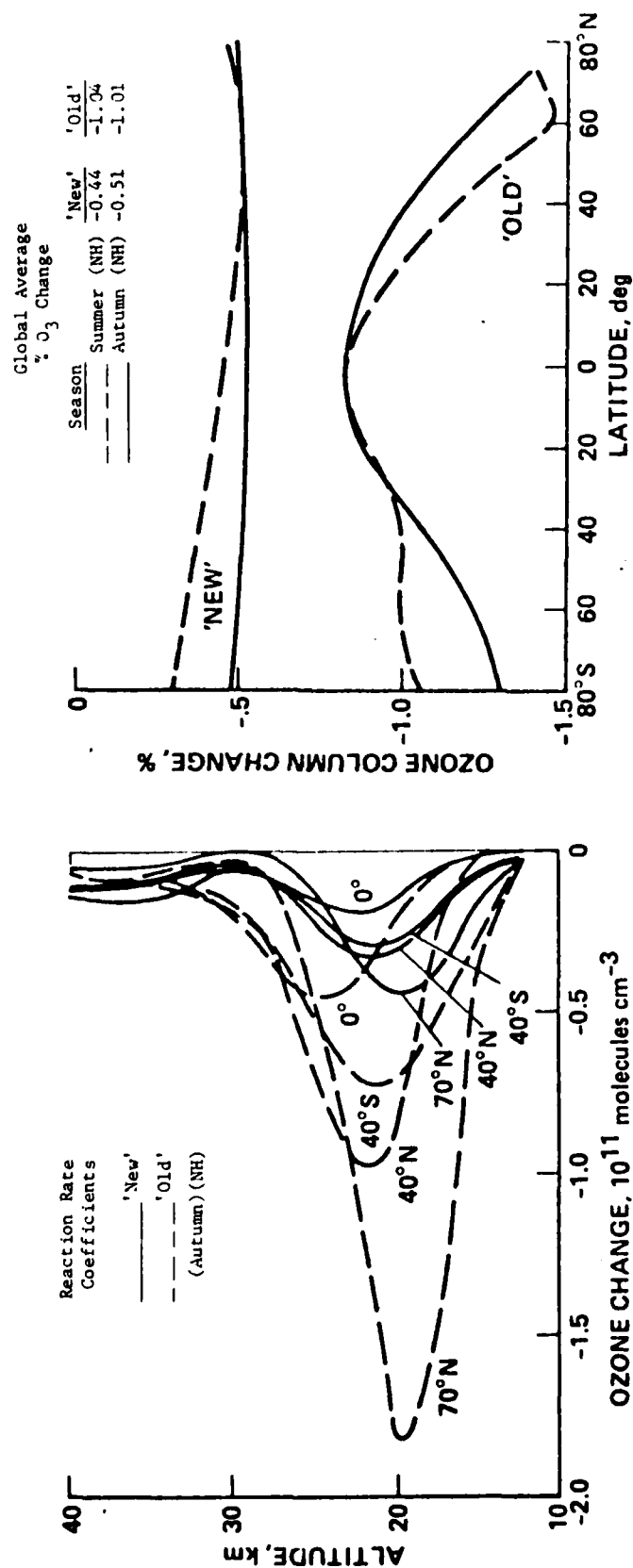


FIGURE 5. 2-D Ozone Change Results Using "Old" and "New" Chemistry Due to  $\text{H}_2\text{O}$  Emissions From Assumed SST Fleet Numbering 250 Flying at 20 km Altitude ( $3.25 \times 10^{10} \text{ kg H}_2\text{O/yr}$ ). (Whitten et al., 1981a)

changes the sign of the ozone effect from positive to negative (Fig. 4) and cuts the negative water effect about in half (Fig. 5). The combined effect is not given. The water effect does not include important thermal feedback effects, and, as noted by Whitten et al., may result in an overestimate of the water effect. This point is discussed in detail in Penner and Luther (1981), who show the reduction in water effects with inclusion of temperature feedback and hydrostatic adjustments.

The latitudinal distribution of effects found by Whitten et al. (1981, 1981a) differs substantially from other 2-D modeling results (see, e.g., Grobecker et al., 1974; Hidalgo and Crutzen, 1977) which have shown much larger differences (twofold to threefold) between the Northern Hemisphere and Southern Hemisphere effects. This is a result of the particular set of transport coefficients used, which are obviously uncertain: Whitten et al. (1981) argue that "Insufficient data are available on the distribution of excess Carbon 14 from nuclear tests and on the ambient distribution of nitrous oxide and methane to permit the transport parameters of one model to be chosen over those of the others." Whitten et al. (1981a) state that the ozone depletion results are similar to those found in CIAP (Grobecker et al., 1974), although for quite different (chemical) reasons. The comparison is indeed somewhat difficult to make, since the CIAP results were adjusted for differences in hemispheric effects which were larger than those reported in Whitten et al. (1981a), and involved an average of 1-D 1974 LLNL and NAS 1975 results. However, comparison can be made to the 1-D 1974 LLNL global average effect (Grobecker et al., 1974, p. E-59) of about 0.75 percent ozone reduction at same injection rate, vs. about 1.3 percent found by Whitten et al. (1981a). The NAS, 1975 model (see National Academy of Sciences, 1975, pp. 116-119; or Oliver et al., 1977, pp. 3-28 to 3-30) using the recommended 2-km altitude adjustment gives 3.24 percent depletion in the Northern Hemisphere. Current numbers are thus within the range of model uncertainty (i.e., variations between models) in CIAP.

The status of current 2-D and 3-D models is given in WMO, 1981. Most current effort continues to be on the halocarbon issue. It is of interest to note that recent 2-D ozone depletion due to halocarbons



calculated by Du Pont have shown almost identical effects in the Northern and Southern Hemisphere (Steed et al., 1981).<sup>\*</sup> However, the halocarbon ozone-perturbing source function is entirely different from that in the aircraft case, as halocarbons are well distributed between the Northern and Southern tropospheres. The Du Pont results also agree reasonably well with 1-D model results. Their 2-D estimate of current depletion caused by CFC11 and 12 ( $\text{CFC1}_3$  and  $\text{CF}_2\text{Cl}_2$ ) is 0.89 percent however vs. 0.50 percent for their 1-D model, implying (to this author) a faster vertical transport in the 2-D model. A slight current asymmetry exists, which would essentially disappear at steady state. Current halocarbon depletion effects, however, are noted to be about balanced by increases in ozone due to  $\text{CO}_2$  increases and aircraft exhaust (citing a paper presented by J.E. Penner at American Geophysical Union meeting, December 1980). This result is apparently consistent with results shown in LLNL, 1981, although a range of values must be recognized as being possible.

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<sup>\*</sup>For continued release of CFC11 and CFC12 at 340,000 and 410,000 metric tons per year, the 2-D model at  $\sim 300$  years model time gives 6.2 percent average depletion vs. 7.0 percent with the 1-D model using identical chemistry. Their model shows 5 to 8 percent depletion at the poles (smaller in the summer) and  $6 \geq 6.5$  percent depletion in the tropics.

## 4.0 OTHER ISSUES AND EFFECTS

### 4.1 1-D EDDY DIFFUSION COEFFICIENTS

The inadequacies of the 1-D  $K_z$  concept are well known;\* the advantages of the concept's simplicity, coupled with the lack of a viable alternative lead to its continued use (and misuse, as when local measurements are compared to "global average" model results). Hence, refinements in  $K_z$  are always of interest. Several recent contributions are noted in Fig. 6. Massie and Hunten (1981) provide a fairly detailed review, and arrive at a curve with eddy diffusivities in the 40-km region about half those commonly used previously. Of more importance to the aircraft problem is the fact that the values assigned near the tropopause are about twofold higher than used in the original Hunten (National Academy of Sciences, 1975) profile. An LLNL "test"  $K_z$  (not "rigorously derived," per LLNL) used by LLNL in attempts to better match the falloff of halocarbon tracers approximates the Massie and Hunten curve at higher ( $> 30$  km) altitudes but is "slower" in the 18- to 25-km region. The Allen et al. (1981) work emphasizes a more complex high-altitude region above that shown in Fig. 6. The value at 50 km used by Allen et al. was constrained to fit the value recommended at this altitude by Hudson (1977), which value appears to be the same as that used in National Academy of Sciences (1976) and (1979).

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\*In this author's view, the 1-D concept applies best to computations involving species such as methane, halocarbons, or nitrous oxide which are well distributed in the troposphere and have sinks in the stratosphere. The concept is also defensible in regions where photochemical equilibrium prevails (i.e., where transport is unimportant). The concept is largely inappropriate for locally-concentrated steady-state sources where seasonal influences are strong, as in subsonics and SSTs near the tropopause; and for pulsed sources where time-dependence is of interest. The concept fails for transport of water vapor. In principle,  $K_z$  varies for each tracer, source, location, and sink.

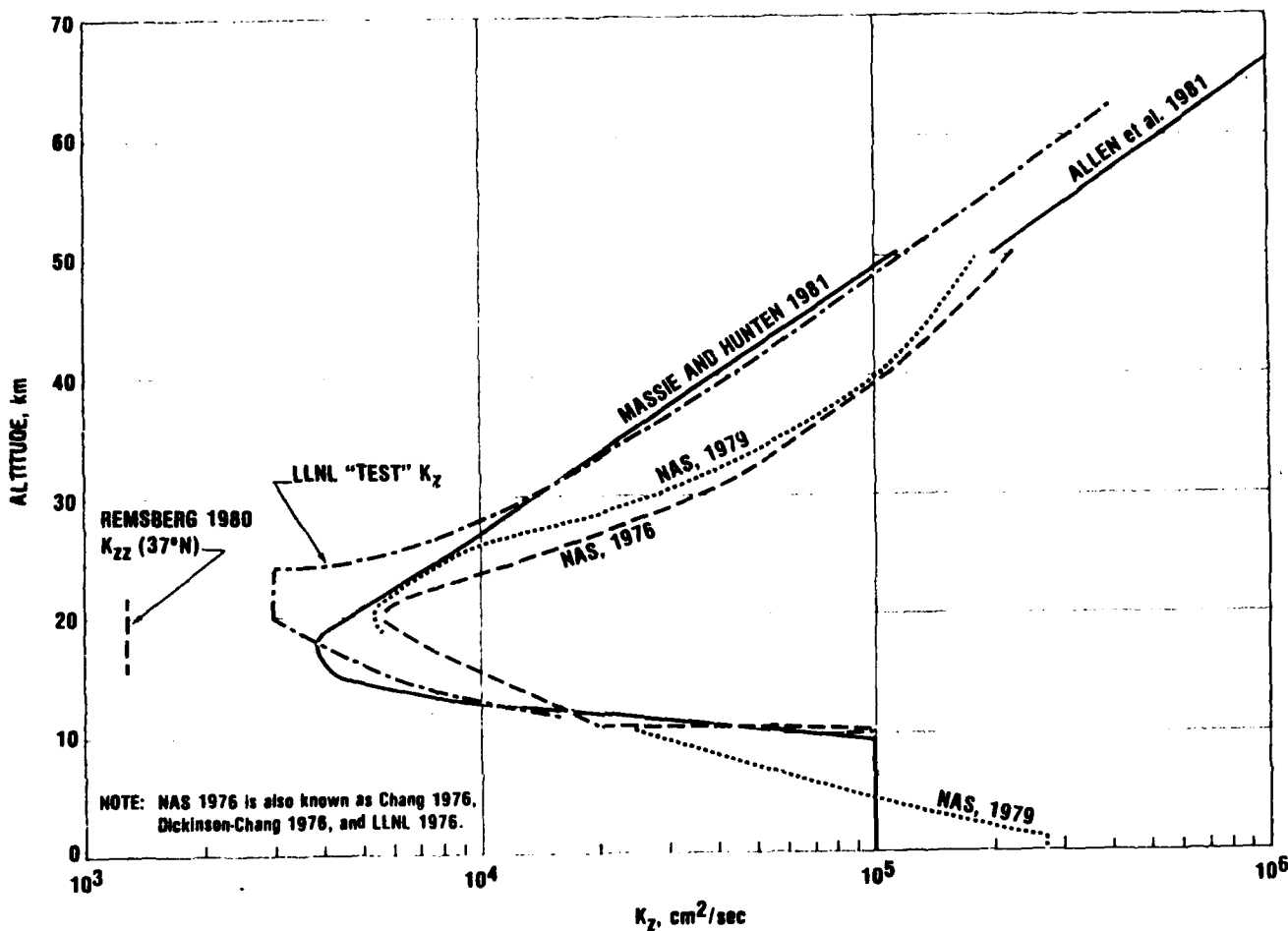


FIGURE 6.  $K_z$  Comparison

Remsberg, 1980, cites smaller  $K_z$  values in the 15-20 km region than shown in Fig. 6. The values were found by studying the vertical spread of a tracer and are presumably lower than found by other techniques due to scale-dependent effects.

The Massey-Huntten 1981 article includes as part of the "composite data set" the  $C^{14}$  treatment used by Johnston et al. (1976). As pointed out by Bauer (1978), this treatment used what appear to us to be inappropriate boundary conditions with a strong sink at the top (tracer concentration at 51 km half that at 50 km, and concentration at a fixed value -- a perfect sink -- at the lower boundary). This sink in the upper stratosphere, coupled with a perfect sink in the troposphere, leads to the need for a "slow"  $K_z$  profile at the tropopause. In any event,

$C^{14}$  is probably a poor tracer because it is not substantially removed on cycling through the troposphere and back into the stratosphere.

## 4.2 MODEL VALIDATION EFFORTS

### 4.2.1 Introduction

A mathematical model of the atmosphere, in which trace species concentrations are computed, e.g., as a function of altitude, can be tested in various ways. Thus, trace species concentrations can be measured and tested against calculated values,\* and reasonable agreement is clearly a prerequisite to any claim of model validity. Unfortunately however, for various reasons, including the fact that models are constructed (to a degree) from such trace species data, it is never clear that the models will correctly predict effects of perturbations. Derwent and Eggleton (1981) have recently discussed this issue with regard to chlorine effects, arguing that trace species data to date are insufficient to distinguish between models giving twofold difference in computed effects. With SST  $NO_x$ , the overall changes are more dramatic, as a netting out of enhancement and depletion regions is involved. Thus, for the SST case, trace species profiles have, with some important exceptions, looked "reasonable" throughout the period shown earlier in Fig. 3. For such reasons, observations of effects following "known" perturbations are of critical importance in assessing the true, or predictive, validity of such models. "Known" perturbations always involve many uncertainties in source strength and altitude, observations involve the usual uncertainties, and pulsed events are not good analogs for continuous sources due to time-dependent transport effects. Nevertheless, where the perturbations can be described reasonably, the model, and particularly its chemistry, can be tested against observations. A number of tests of this type have been attempted and are described in Section 4.2.2.

### 4.2.2 Nuclear Weapons Tests

The first test of the predictive, although post hoc, validity of the ozone models (showing depletion from  $NO_x$  injections into the stratosphere)

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\*Difficulties obviously exist, in that measurements are always localized in time and place, whereas a model may purport to give a "global" or "zonal" mean value.

was the comparison of ozone data to model calculations following the atmospheric weapons tests of the 1955-1967 period.

These tests put large amounts of  $\text{NO}_x$  into the stratosphere at altitudes of 15 to 25 km, crudely similar to the perturbations which might be expected from an SST fleet, and therefore provided much interest.

The topic has been reviewed by a number of authors (Foley and Ruderman, 1972, 1973; Johnston et al., 1973; Bauer and Gilmore, 1975; Luther, 1977; Bauer, 1979; Chang et al., 1979). It is worth noting that the estimated maximum 1 to 3 percent depletions in the Northern Hemisphere calculated using 1979 chemistry (Bauer, 1979) seemed more easily acceptable superficially as being within the noise of the ozone data than were the earlier 5 to 8 percent reduction estimates (Luther, 1977); these larger values would presumably again be computed using present chemistry. (As far as is known, the calculations have not been repeated.) In a recently published paper (Reinsel, 1981), based on time series modeling of monthly ozone data over the period 1958-1979, it is stated that the ozone data "are consistent with a maximum decrease in total ozone in the Northern Hemisphere of approximately 2 to 4.5 percent due to nuclear testing effects in the early 1960's. Little evidence of any change ( $0.49 \pm 1.35$  percent) was found for the 1970-1979 period. Continued study of these events seems to be needed, particularly with 2-D or 3-D models.

#### 4.2.3 The Solar Proton Event of August 1972

The solar proton event (SPE) of August 1972 was the largest in 25 years (and since). The event introduced large quantities of  $\text{HO}_x$  and  $\text{NO}_x$  above 10 mbar (31 km) poleward of about  $55^\circ$  latitude. This is a region where ozone chemistry reequilibrates rapidly following perturbations, and transport processes are relatively unimportant. Nimbus 4 showed a sudden drop in ozone above about 4 mbar (38 km), the region in which most of the energy was deposited. A number of papers resulted (Heath et al., 1977; Fabian et al., 1979; Jackman et al., 1979) which were reviewed very briefly in Oliver (1979). In general, it can be said that the abrupt drop-off noted above 4 mbar compared well to model calculations, but the time-dependent response thereafter was somewhat unsatisfactory. Furthermore, there were, and are, a number of uncertainties involved, and in the

principal early paper the chemistry used in the modeling (Heath et al., 1977), is now known to have been incorrect.

This event has been reanalyzed in two recent papers (Solomon and Crutzen, 1981; Reagan et al., 1981). The first of these emphasizes effects of recent chemistry changes, including chlorine species, and presents calculated effects from  $\text{HO}_x$  and  $\text{NO}_x$ ; the second emphasizes observations, but also includes calculated  $\text{HO}_x$  and  $\text{NO}_x$  effects, although chlorine species were not included in the model.

The Solomon and Crutzen (1981) article includes revised calculations, including a thermal feedback effect (cooling by  $6^\circ$  at 45 km 8 days after the event) and revised observations, which together considerably improve the correspondence. The 7+ percent depletion "observed" at 30 km previously has now disappeared, in line with model results. Also, the observed effect extends upward to 55 km instead of 50 km, as previously, again, in line with models. The current results are based on 1-D models, although the Crutzen 2-D model was used earlier (Heath et al., 1977). The authors find that inclusion of  $\text{ClO}_x$  is important to the calculations and, surprisingly, that use of the fast  $\text{HO}_2 + \text{NO}$  rate (40 times that used previously) actually increases ozone depletion from the SPE above about 35 km, contrary to its effect at lower altitudes of 17 to 20 km. Inclusion of temperature feedback effects is also important. Solomon and Crutzen find that this SPE provided an unusual opportunity for evaluation of  $\text{NO}_x$  effects at these altitudes.

In Reagan et al. (1981), the event and response information are described in detail. The event lasted some 10 hrs in its most intense period, at the end of which the ozone concentrations at latitudes 75 to  $79^\circ\text{N}$  were reduced by 46, 16, and 4 percent at 49.5, 41, and 32 km altitudes, respectively, with about 2 percent reduction at ground level. The polar ozone cavity was observed to rotate in a "semi-rigid mass" for 53 days, at which time wind changes prevented further tracking. These decreases resulted in very large increases in ambient  $\text{NO}_x$  (a factor of 45 at 50 km, 15 at 45 km, and 4 at 40 km). Reagan et al. also note the effects on local temperatures, and give an unusually good description of some of the uncertainties and difficulties involved.

As noted, SPE events and their analyses provide an opportunity for study of  $\text{NO}_x$  and  $\text{HO}_x$  effects at altitudes where equilibrium photochemistry dominates. Use of a 1-D model to study effects in a "semi-rigid mass" circling the pole would hardly be justified were this not the case. Even so, the results must be interpreted with some care. The event and its analyses provide valuable confirmation of  $\text{NO}_x$  effects at high altitudes, regarding the SST problem.

#### 4.2.4 The Tunguska Meteorite Event of 1908

Turco et al. (1981) have analyzed the effects on ozone of what is presumed to have been a meteorite (a "small" cometary fragment) which broke up in the air over Siberia ( $61^\circ\text{N}$ ,  $102^\circ\text{E}$ ) with the force of a "10-megaton nuclear detonation" on 30 June 1908. The event has long been of speculative origin.

Turco et al. suggest the event injected some 30,000,000 metric tons of NO into the atmosphere between 10 and 100 km, which they state to be roughly 5 times the amount of odd nitrogen ( $\text{NO}_x = \text{NO} + \text{NO}_2 + \text{HNO}_3$ ) in the entire earth's stratosphere. Turco et al. calculated, using a 1-D model, that this amount of NO would deplete ozone by about 45 percent in the first year on a hemispheric average, figures comparable to those computed for a nuclear war. Column recovery (to about 90 percent levels) required about 5 yrs. If deposition was assumed to be in the  $55$  to  $65^\circ\text{N}$  zone, column depletions above 10 km of more than 85 percent were calculated. Turco et al. then examine ozone column measurements deduced from Mount Wilson transmission data from 1909-1911. The means of the data matched the calculated ozone curve reasonably well, although the very wide scatter in measured column ozone (e.g., in 1910 from 0.17 to  $1.27 \times 10^{19}$  molecules/cm<sup>2</sup>) make the correlation somewhat unconvincing.

These calculations, while involving many uncertainties, are of considerable interest in terms of climatic and ecological effects, or lack thereof. Turco et al. examine the 1909-1911 period for climatic anomalies, of which there were a number, including reduced Northern Hemisphere temperature, fewer tropical cyclones, etc. Volcanic eruptions in 1907 and 1912 may well have contributed to, or been dominant in creating, such effects. (See Oliver, 1976.) It would be interesting to examine any data

on this period for effects on various organisms of increased erythermal ultraviolet (doubled for 3 yrs) as using data, for example, on wheat or fishery yields. It is also important to note, as do Turco et al., that in spite of these massive perturbations, no drastic climate change was triggered and that weather over much of the globe was not affected in any obvious way.

#### 4.2.5 The NASA Confirmation

In mid-August 1981, NASA (see Norman, 1981) released carefully qualified statements which suggested that observed ozone depletions in the region of the atmosphere near 40 km seemed to be matching model predictions. More details are provided on this point in an abstract by Heath and Schlesinger (1981) of a paper planned for presentation at the December 1981 meeting of the American Geophysical Union (AGU). Heath and Schlesinger report on evidence of a 0.5 percent/yr ozone decrease between 1970 and 1979 near 40 km. They state "At present, a likely explanation for the observed decrease is due to the predicted effects of chlorofluorocarbons." They also note that "An apparent solar cycle variation of ozone has been found which parallels that from Umkehr observations but is contradicted by current assumptions of UV solar variability and 1-D models." Note that this is a local and not a full column, and as such does not contradict the observations of Reinsel (1981) discussed earlier, which were for the full ozone column. (See Section 3.2.) This partial confirmation of the chlorine theory is an important one, and will be given close examination. Causation, in the absence of chlorine measurements, cannot be unequivocally claimed, as the NASA authors are well aware.

#### 4.3 SKIN CANCER INCIDENCE AND OTHER UV EFFECTS

The skin cancer issue is associated with ozone depletion, because such depletion increases the erythermal dose rate (Cutchis, 1974 and 1978). No new studies of the effect of aircraft emissions on skin cancer incidence have been undertaken. Part of the reason, of course, is that over the 1977 to 1980 period both subsonic and supersonic aircraft NO<sub>x</sub> emissions were computed to increase the ozone column. This situation still applies in practical terms; the few operating Concorde can have no significant



effect on the ozone enhancement expected from the subsonic fleet. This enhancement, coupled with build-up in CO<sub>2</sub> has, as reported earlier, been computed to about balance the computed depletion at higher altitudes by halocarbons from 1970 to 1980, at least in a 1-D sense.

Several items may be noted. The first is a news item (Altman, 1979) which mentions documentation of increased skin cancer risk for patients undergoing a relatively new (since 1974) treatment for psoriasis with ultraviolet light, plus a drug. The statistics cited (such as that patients undergoing the treatment have a 2.63 times greater probability of developing skin cancer than a group of persons without psoriasis living in the same general area) are not presented in terms of doses given. The skin cancers found were squamous cell carcinomas, which are treated by excision. These observations are in at least qualitative accord with much earlier information attributing non-melanoma skin cancers to ultraviolet light (National Academy of Sciences, 1975, 1976, and 1979; Cutchis, 1978; Maugh, 1980).

Smith and Baker (1980) discuss changes in marine productivity with changes in stratospheric ozone using a Carbon-14 short-term incubation technique, which they conclude is inadequate for possible large amplification factor processes that may be ecologically significant. Since the majority of the photoinhibitive effect they discuss is at wavelengths above the UV-B region and not affected by ozone depletion, there should be little such effect due to UV-B changes. Gerstl et al. (1981) discuss the amplification factor in terms of changes in biologically damaging radiation with depletions in ozone. The ozone depletions are assumed to be caused by chlorofluorocarbons, with latitudinal distributions found by a 2-D model. They find this factor to be between 1.9 and 2.2 for erythermal weighting, and between 2.5 and 2.8 for DNA weighting. A factor of 2 has often been used in the past with erythermal weighting, but the range of uncertainty was wider (Gerstl et al., 1981). As shown by Cutchis (1974), this factor changes with degree of ozone depletion. Use of such data to calculate changes in skin cancer rates involves further uncertainties (Cutchis, 1978).

A related subject of interest is discussed by Zardecki and Wiser (1981)(abstract only available at time this paper is being written), who point out the fact that tropospheric aerosols reduce UV flux at the ground. As an example, they note that a 5 percent ozone decrease increases solar UV flux by 12 percent, while the aerosol content which corresponds to a 25 km surface visual range decreases UV flux by the same amount relative to clean air. A similar "compensation" applies to erythemally weighted UV flux. Such effects have been cited in earlier studies (e.g., Mo and Green, 1975; Cutchis, 1980). It should be noted that compensation implies simultaneity of events in time and place and is relative to some ideal preexisting condition; the significance is thus unclear.

#### 4.4 CONTRAIL EFFECTS

In a recent study, Changnon et al. (1980) have examined the effect of contrail cirrus on surface weather conditions in the Midwest. While the study is carefully caveated in view of various uncertainties and the ever-present correlation-causation question, the study strongly suggests\* that regions experiencing heavy and increasing jet traffic are also areas with increased numbers of cloudy days (and thus a decreased number of sunny days), moderated temperatures (a smaller difference between daytime maximum and nighttime minimum) and probably (p. 53 their report) greater precipitation. The authors point out that these regions of heavy jet traffic are also regions of heavy industrialization, which confuses cause-effect relationships, particularly in matters such as haziness.

The region studied is shown in Fig. 7; one of their plots of results is shown in Fig. 8. The authors note (as in Fig. 8) that apparent trends also existed at times over certain periods long before jet aircraft became commonplace. However, since increased high cloudiness in regions of

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\*To quote p. 47, "Stations along the east-west line where contrail production is greatest ... all showed a notable increase in moderated months in 1960-70, the period when jet aircraft significantly increased ... Since the increased cloudiness in the 1961-77 period may likely be a result of the contrail-induced cirrus in the central area, it appears the effect has at least partially caused a moderation of temperatures."



FIGURE 7. Stations and Areas Used by Sky Cover and Sunshine Studies. (Changnon et al., 1980)

heavy jet traffic had been observed previously (see Figs. 9 and 10, Machta, 1971) and since contrail formation under appropriate conditions is undeniable, the Changnon et al. findings do not seem unreasonable. Contrail effects are largely a local or at best regional phenomenon, and very high traffic densities are required to make even a moderate impact. Some quantification of this statement is desirable and is provided below after some background discussion.

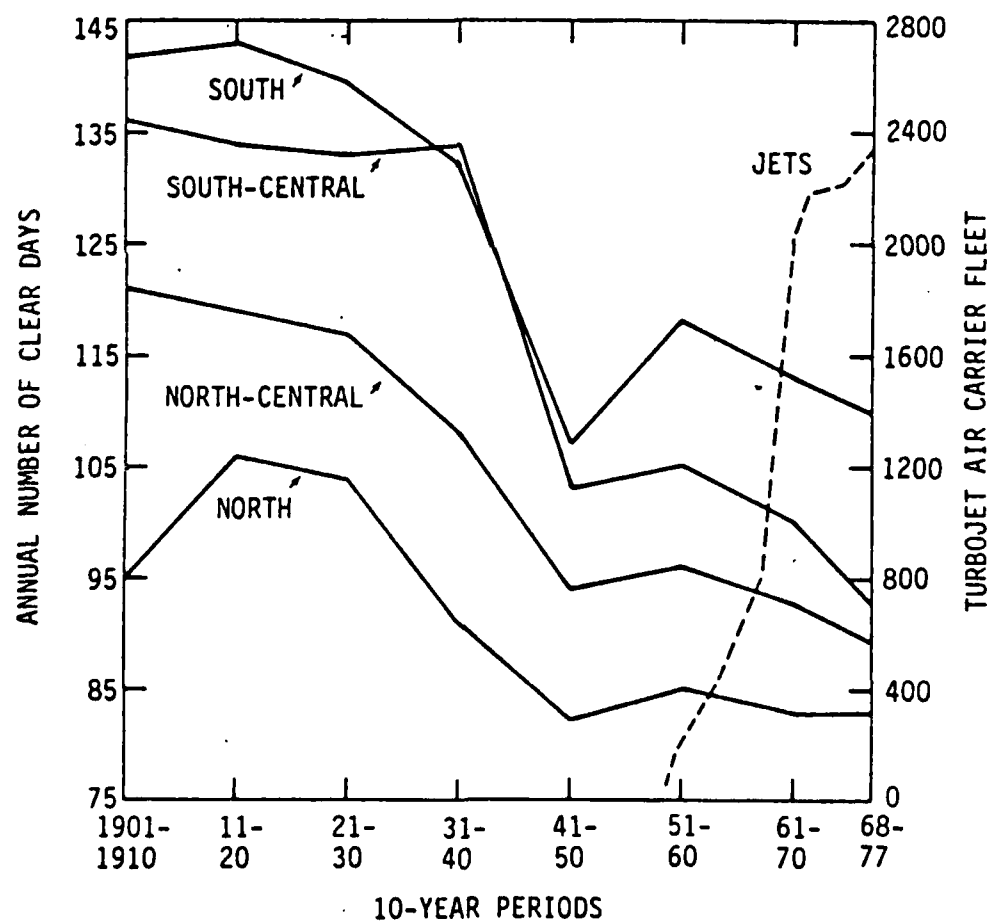


FIGURE 8. Annual Clear Days, 1901-1977, Per Year Means, as Reported by Changnon et al., 1980.

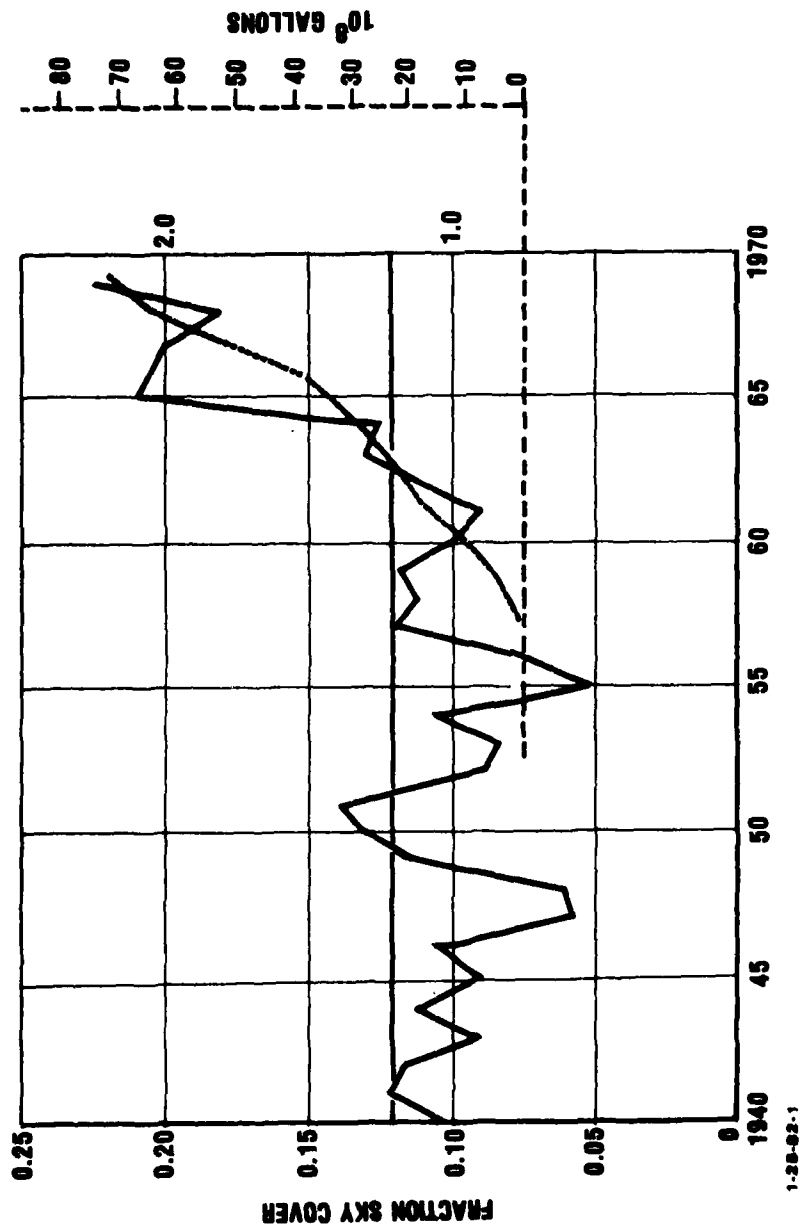
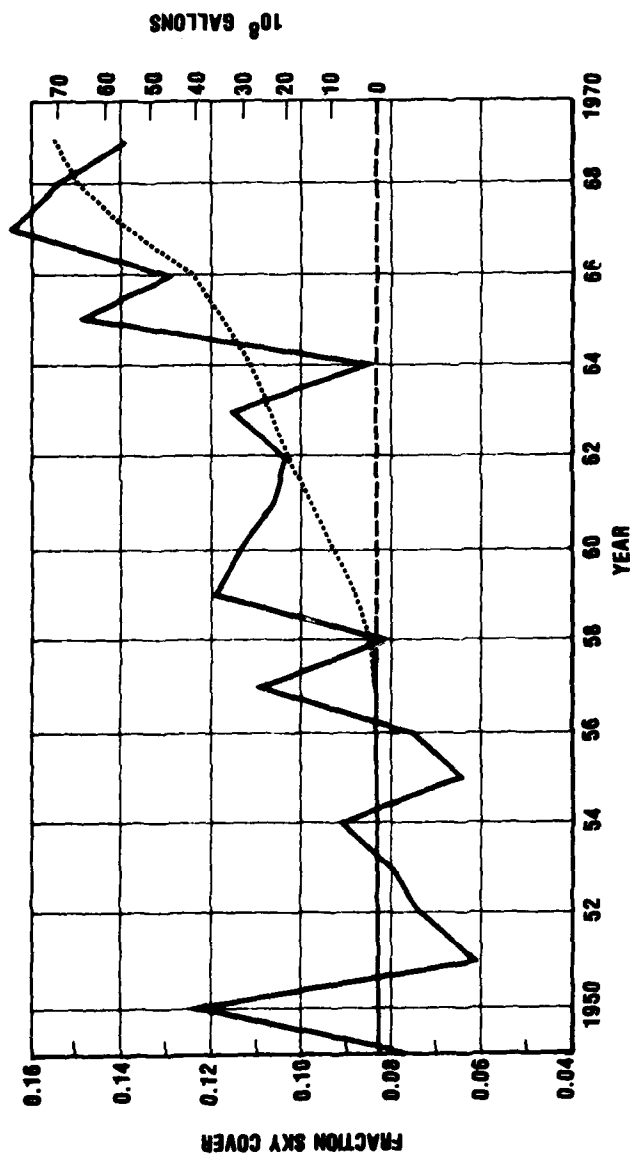


FIGURE 9. Annual amount of high cloudiness at Denver, Colorado, with no low or middle clouds. Data obtained from three hourly Weather Bureau airways observations. The dashed curve represents the annual domestic commercial jet fuel consumption for the United States which is assumed to be proportional to the number of flights in the vicinity of Denver. Source: Machta and Carpenter, 1971.



Source: T. Carpenter, Met. Statistics, ESSA.  
1-28-82-2

FIGURE 10. Annual amount of high cloudiness at Salt Lake City, Utah, with no low or middle clouds. Data obtained from three hourly Weather Bureau airways observations. The dashed curve represents the annual domestic commercial jet fuel consumption for the United States, which is assumed to be proportional to the number of flights in the vicinity of Salt Lake City. Source: Machta and Carpenter, 1971.

Contrail formation and contrail effects have been discussed for many years (e.g., Appleman, 1957, 1965; Machta, 1971, 1971a; Machta and Telegadas, 1974; Oliver, 1970; SCEP, 1970; Kuhn, 1970; Beckwith, 1972; Knollenberg, 1972; Joseph et al., 1975; Appleman and Barrett, 1975). The conditions under which contrails can form are well known. Formation of contrails in the stratosphere is infrequent, due to the combination of low relative humidity and temperature. Carrying passengers by SST, rather than by subsonic aircraft, would reduce contrail frequency. The principal questions about contrail effects relate to their persistence and frequency, and where persistent (since persistence demands near- or super-saturation) whether formation of cirrus is simply accelerated (triggered) by aircraft passage, and would shortly have occurred in any event. Knollenberg (1972) has pointed out that persistent contrails contain as much as  $10^4$  times the water content per unit of length as that added by the aircraft; furthermore, the resultant hydrometeors, by settling, cause a drier atmosphere than existed before aircraft passage. Kuhn (1970) showed that persistent contrails would reduce temperatures under such contrails by  $5.3^\circ\text{C}$ ; 5 percent persistence would lead to a  $0.15^\circ\text{C}$  drop. These results, which are not explicitly indicated to be referring to 24-hour averages (cooling would be expected under a contrail on an otherwise sunny day), seem to disagree with modeling results in the COMESA study, where increases in high "half-black" cirrus (over cloudless) caused a global average warming\* of  $0.86\text{ K}$  for 22.6 percent coverage. "Full black" cirrus cause greater warming. Low clouds had a much greater (and cooling) effect.

The climatic effects of aircraft contrails on a global basis can only be estimated on an extremely crude basis, because frequency and coverage data are lacking. As an exercise, however, if 10,000 subsonic aircraft were each to travel 1000 km/hr, 8 hours per day, each generating a contrail 1 km wide and lasting 1 hr, then  $3 \times 10^6\text{ km}^2$  of contrails would exist continuously, which represents 0.6 percent of the earth's area and would, ratioing the COMESA results, cause warming of  $0.3\text{ K}$ . However, per

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\*Similar earlier results from modeling work by Manabe and Weatherald (1967) are cited by Machta and Telegades (1974).

Beckwith, only 7 percent of all observations involved persistent (> 5 minutes) contrails under otherwise clear sky or scattered cloud conditions (25 percent were found to be persistent, but the majority of these occurred with existing cloud decks), and many of these did not last 1 hour; the expected global average warming effect would be of the order of 0.001 K or less. This, of course, does not preclude local effects in regions of high traffic, nor does it preclude contrails having a cooling effect (under otherwise clear sky conditions) during the day, as found by Kuhn (1970).

One final comment on the Changnon et al. (1980) results. Moderations in day-night temperature differences, with reduced peak temperatures, may well be beneficial to agriculture, as is increased rainfall, although the actual timing and magnitude of the changes makes any evaluation of effects on agriculture very complex (Bartholic et al., 1975).

#### 4.5 GENERAL CLIMATIC EFFECTS

Lacis et al., 1981 have examined the effects of certain trace gas increases between 1970 and 1980. The model derived equilibrium greenhouse warming. The increases in concentrations used are presented in Table 14.

TABLE 14. CLIMATIC CHANGE FIGURES

	<u>Arbitrary Change</u>			<u>1970-1980 Change</u>		
	<u>a<sub>0</sub> (ppb)</u>	<u>Δa<sub>0</sub> (ppb)</u>	<u>ΔTeq (°C)</u>	<u>a<sub>0</sub> (ppb)</u>	<u>Δa (ppb)</u>	<u>ΔTeq (°C)</u>
CH <sub>4</sub>	1600	1600	0.26	1500	150	0.032
N <sub>2</sub> O	280	280	0.65	295	6	0.016
CCl <sub>3</sub> F	0	2	0.29	0.095	0.135	0.020
CCl <sub>2</sub> F <sub>2</sub>	0	2	0.36	0.125	0.190	0.034
CO <sub>2</sub>	300,000	300,000	2.9	325,000	12,000	0.14

a<sub>0</sub> = Initial condition.

ΔTeq = Change in equilibrium temperature.



Note that the sum of the non-CO<sub>2</sub> effects considered is 0.10 °C, and that due to CO<sub>2</sub> is 0.14 °C. These changes, of course, are not all the changes which occurred in this period, as the authors note. A figure (-0.02 °C) is quoted for the 5 percent ozone decrease (0.5 percent) per year for 10 years) at 30 to 50 km suggested by D.F. Heath (Heath and Schlesinger, 1981; Section 4.2.5). Actual warmings would be somewhat less due to thermal lag (perhaps 6 year characteristic time) in the oceanic mixed layer, Lacis et al. (1981) conclude that greenhouse-effect warming in the range 0.1 to 0.2 °C occurred in the 1970s. Note that the authors did not include any effect from upper tropospheric ozone increases due to aircraft NO<sub>x</sub>, which, theoretically, would cause warming (Fishman et al., 1979), of magnitude which would need consideration in a tabulation such as in Table 13,\* nor any of many other postulated climatic effects (stratospheric water changes, albedo changes due to desertification or deforestation, solar-cycle effects, etc.) so that the assessment is incomplete.

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\*Fishman et al. (1979) find that a doubling of tropospheric ozone would increase surface temperature by 0.9 K. Lack of current modeling results make it impossible to tie this directly to aircraft effects, but judging from earlier results, a 10 percent increase in upper tropospheric ozone (1 percent increase in column over the pre-jet aircraft period) would not be unreasonable by 1990. The corresponding increase, if linear, would be 0.09 K.

## 5.0 CLOSING COMMENTS

The foregoing discussion makes clear the dynamic nature of investigations into the effects that man's various activities may have on the ozone layer and on climate. The many changes in calculated effects on the ozone layer over the past decade have made clear the difficulties involved in this complex subject, but should be viewed positively: it may be that a reasonably coherent picture is emerging. Certainly much dramatic progress has been made, but just as clearly, much remains to be done.

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